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Generation and propagation of polarization singularities in second harmonic generation from the surface of the isotropic gyrotropic medium

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Abstract

When proceeding to the oblique incidence from the normal incidence geometry in surface second harmonic generation, the symmetry of the problem and, thus, of the reflected signal beam breaks. In this case there can appear C-points in the transversal section of the signal beam. Using the analytical formulae obtained in previous works, we studied the conditions of appearance and the behaviour of the C-points in reflected signal beam (in particular case of SHG), and also the way of the line-to-point singularity transformation.

Introduction

Polarization singularities in paraxial light beams are lines or surfaces in the propagating beam where the intensity of one of the orthogonally (circularly or linearly) polarized components of the radiation becomes zero. These objects have been thoroughly investigated in a big number of theoretical and experimental works in linear optics area (see [1 – 12]). In one of the first papers on the polarization singularities there have been defined terms C-lines or L-surfaces which are geometric places of the points in the propagating beam where the radiation is circularly or linearly polarized. When considered in the cross-section of the propagating beam polarization singularities are treated as C-points and L-lines (as the intersections of lines or surfaces with the transversal plane). Unlike the conventional optical vortices (or screw dislocations) these singularities can be considered as "component" optical vortices. In the vicinity of the C-points there can appear generally three morphological types of the electric field distribution called "star", "lemon" and "monstar" (see [1-6]).

Later the conditions of appearance and the behavioral features of the polarization singularities have been studied in different problems of linear optics, such as evolution of the random vector fields in isotropic media [2], propagation of laser radiation in birefringent chiral crystals [3], polarization singularities in sky in daylight [4], propagation of optical vortices in birefringent crystals [10,11], coherent interaction of orthogonally polarized Bessel beams [12]. The fundamental interconnection of the anisotropy of Stokes parameters in the vicinity of the C-points with the morphology of the last ones have been established [5]. Mechanisms leading to the appearance of the polarization singularities in beams with initially regular intensity and polarization distribution propagating in inhomogeneous media, waveguides and laser resonators were intensively studied. Highly-efficient experimental methods of detection of polarization and phase singularities in light beams have been developed [7 – 10]. Paper [6] gives review of these and other relevant works. In spite of the thorough investigations of polarization singularities in a broad range of problems of linear optics there exists only a very limited number of works concerning the appearance and the evolution of polarization singularities in nonlinear optical processes.

In earlier works [13] the analytical formulae were obtained, which wholly describe the signal beam at sum-frequency generated from the surface of the isotropic chiral medium (spatial symmetry group $\infty\infty$) by oblique incident elliptically polarized Gaussian profile fundamental beams in the case of arbitrary (non-coplanar) geometry of incidence. The spatial dispersion of the quadratic optical response of the medium and the peculiar features of medium surface response (compared to that of medium bulk) have been taken into account by means of modified boundary conditions [14,15]. It was shown that the polarization state of light changes along the transversal section of the reflected beam at sum-frequency, and for typical values of the angles of incidence (not too small, like $\sim 1°$, and not too big, like $> 70°$) and small divergence angles of the incident beams the polarization state variations are minor within the central bright area of the signal beam. Otherwise, for big and small values of the angles of incidence the polarization alternations along the beam cross-section become very strong. In the last case the intensity distribution is not Gaussian any more.

In [16] there has been found the coordinate dependence of the electric field of the second harmonic beam reflected perpendicularly to the surface in the abovementioned problem. The appearance of the double frequency signal in case of normal incidence of the fundamental is forbidden within the framework of the plane-wave approximation theory [17], however, accounting for the focusing of the incident beam and small longitudinal component of the electric field in it, the appearance of the reflected second harmonic beam in this case can be explained. It was found that in this geometry of interaction the signal beam has non-Gaussian profile and the polarization is distributed inhmogeneously in its cross-section, while the incident beam is uniformly polarized and has Gaussian profile. The structure of the signal beam possesses a kind of symmetry, such, that the polarization state in the transversal section of the beam depends only on the polar angle coordinate and does not depend on the distance from the center of the beam: polarization remains the same along any straight line crossing the center of the beam. This symmetry makes impossible observation any point singularities (like C-points) in perpendicularly reflected second harmonic beam. Instead of them one can observe C-lines (lines of circular polarization) and L-lines, crossing the center of the beam (where the intensity of light is zero). The conditions of the appearance of these singularities and analytical expressions for the polar angles determining their position in the beam cross-section can be easily found from the formulas given in [16] which represent itself particular case of the formulae in [13]. It is remarkable that the beams with analogue structure appear in problems studied in [18, 19].

Present work is devoted to the studies of the conditions of appearance and behaviour of the C-points in the cross-section of the double frequency beam generated from the planar surface of the isotropic chiral medium with spatial dispersion of quadratic nonlinearity by oblique incident elliptically polarized Gaussian profile beam at fundamental frequency. Apart of this we examine the behaviour of the C-lines and the C-points and the signal beam structure transformation, when proceeding from the normal incidence geometry to the oblique one (at zero and small values of angle of incidence).

Main equations. Formulation of the problem.

We start our analysis from the expression for the electric field in the sum-frequency beam generated from the surface of the isotropic chiral medium by two oblique incident elliptically polarized Gaussian beams at fundamental frequencies [13]. In this case the response of the nonlinear medium is represented by the local and non-local nonlinear bulk contribution and by the nonlinear surface current density contribution [14,15]. All the contributions are proportional to the product of the electric fields of the incident beams and they represent themselves the origins of the sum-frequency radiation. The amplitude and the polarization of the reflected wave at sum-frequency can be found from the modified boundary conditions [14, 15]. The last ones account for the difference between the properties of the thin layer near the medium surface and ones of the medium bulk, particularly the difference between the symmetry properties.

Using the abovementioned expressions from [13] it is not difficult to deduce the formula for the coordinate dependence of the electric field in reflected beam at double frequency, when one elliptically polarized monochromatic Gaussian beam falls onto the medium surface at θ_1 angle of incidence with the electric field $\mathbf{E}_1$:

$$\begin{aligned}&\mathbf{E}_1(x_1, y_1, z_1, t) = \left(\mathbf{e}_1 + \frac{\mathrm{i}\mathbf{e}_z^{(1)}(\mathbf{e}_1\cdot\nabla)}{k}\right)\times\\&\times\frac{E_0}{\beta(z_1)}\exp\left\{-\frac{x_1^2+y_1^2}{w^2\beta(z_1)} - \mathrm{i}\omega t + \mathrm{i}kz_1\right\}\end{aligned} \qquad (1)$$

Here the E_0 is the amplitude, ω is the frequency and w is the effective waist size of the incident beam, c is the light velocity in vacuum, $\lambda = 2\pi c/\omega = 2\pi/k$ is the wavelength, $\beta(z_1) = 1 + 2\mathrm{i}z_1/(kw^2)$, $\mathbf{e}_z^{(1)}$ is the unit vector directed along the z_1-axis of the $x_1y_1z_1$-coordinate system, associated with the incident beam (where x_1 lies within the plane of incidence). Complex polarization vector perpendicular to $\mathbf{e}_z^{(1)}$ is defined as the following

$$\begin{aligned}\mathbf{e}_1 &= \sqrt{(1-M_1)/2}\,\exp(-\mathrm{i}\Psi_1)(\mathbf{e}_x^{(1)} + \mathrm{i}\,\mathbf{e}_y^{(1)}) +\\&+\sqrt{(1+M_1)/2}\,\exp(\mathrm{i}\Psi_1)(\mathbf{e}_x^{(1)} - \mathrm{i}\,\mathbf{e}_y^{(1)}).\end{aligned} \qquad (2)$$

Here $\mathbf{e}_{x,y}^{(1)}$ are the unit vectors directed along the x_1 and y_1 axes, $\Psi_1 = 0.5\arg\{E_{1+}E_{1-}^*\}$ is the angle of orientation of the polarization ellipse, $M_1 = (|E_{1+}|^2 - |E_{1-}|^2)/(|E_{1+}|^2 + |E_{1-}|^2)$ is the ellipticity degree of the polarization ellipse of the incident light, $E_{1\pm} = E_{1x_1} \pm iE_{1y_1}$ are the circularly polarized components of the light field. Parameter M_1 changes between -1 (circular polarization with counterclockwise rotation) and 1 (circular polarization with clockwise rotation) through the 0 (linear polarization), while Ψ_1 changes from 0 to π (polarization states with $\Psi_1 = \psi_0$ and $\Psi_1 = \psi_0 + \pi$ are undistinguishable). It is easy to show that $|\mathbf{e}_1|^2 = 1$. We remark especially that (1) contains also the longitudinal component of the electric field, which is necessary in order to satisfy Maxwell equation $\operatorname{div}\mathbf{E}_1 = 0$ in vacuum. Electric field distribution $\mathbf{E}_1$ satisfies this condition within the first order approximation on the divergence angle of the beam, i.e. when $w >> \lambda$.

The beam at frequency 2ω will be reflected at the angle θ_1 relatively to the perpendicular to the surface which is equal to the angle of incidence of the fundamental beam. The symmetry group of the isotropic gyrotropic medium is $\infty\infty$ (surface symmetry group is ∞), therefore the local quadratic optical response of the medium bulk described by the $\hat{\chi}^{(2)}(2\omega;\omega,\omega)$ is absent. In xyz-coordinate system which is connected with the reflected double frequency beam (z-axis coincides with the direction of propagation) the formula for the transversal component $\mathbf{E}_\perp$ of the electric field vector in this beam can be deduced from [13]:

$$\mathbf{E}_\perp(x,y,z,t) = \mathbf{E}_\perp^{(G)} + \mathbf{E}_\perp^{(NG)} = \frac{\pi^2 E_0^2 \cos\theta_1}{(1+\mathrm{i}z/l)} \times$$
$$\times \exp\left(-\frac{2(x^2+y^2)}{w^2(1+\mathrm{i}z/l)} - 2\mathrm{i}\omega t + 2\mathrm{i}kz \right) \times \tag{3}$$
$$\times \Big[(a_{3x}\mathbf{e}_x^{(2)} + a_{3y}\mathbf{e}_y^{(2)}) +$$
$$+ \left(\frac{\tilde{a}_{3xx}x + \tilde{a}_{3yx}y}{(1+\mathrm{i}z/l)}\mathbf{e}_x^{(2)} + \frac{\tilde{a}_{3xy}x + \tilde{a}_{3yy}y}{(1+\mathrm{i}z/l)}\mathbf{e}_y^{(2)} \right) \Big] . =$$

Here $l = kw^2/2$, $\mathbf{e}_x^{(2)}$ and $\mathbf{e}_y^{(2)}$ are the unit vectors along the x- and y-axes (the x-axis lies in the plane of incidence). Cumbersome expressions for the complex-valued coefficients a_{3x}, a_{3y}, $\tilde{a}_{3xx}$, $\tilde{a}_{3xy}$, $\tilde{a}_{3yx}$, $\tilde{a}_{3yy}$ are not given in this text since they are not really relevant to the subject of the present work. These coefficients depend on the polarization state of the incident beam, the angle of incidence θ, medium refraction indices n_ω and $n_{2\omega}$ at frequencies ω and 2ω, the components of the tensors $\gamma_{ijkl}^{(2)}$ and $\kappa_{ijk}^{(2)}$, standing for the nonlocal quadratic medium response and nonlinear surface current density response in the modified boundary conditions in the medium with $\infty\infty$ symmetry. Information on the possible numerical values of the components of these tensors as well as brief review of the experimental works where they were measured can be found in [13,16].

Discussion of results.

When considering two terms in square brackets in (3) it should be noted that for typical values of the nonlinear susceptibilities of the medium and for $5° \le \theta_1 \le 75°$, the following relation is true: $|\tilde{a}_{jk} w / a_j| \sim \lambda / w$ (where j,k can take values "x", "y"). Therefore, in the central bright part of the signal beam (where $x, y \le w$) for the incident beams with small divergence angles (i.e., $\lambda / w \le 0.1$) the second term in the brackets in (3) responsible for the $\mathbf{E}_\perp^{(NG)}$ (non-Gaussian part of the field) is much smaller than the first one. In this case the beam at double frequency will be almost uniformly polarized in its central part and the shape of its intensity profile will be very close to Gaussian. Though, $\mathbf{E}_\perp^{(NG)}$ can exceed $\mathbf{E}_\perp^{(G)}$ (or can become comparable with it) for $\theta_1 \ge 75°$ or $\theta_1 \le 5°$. In this case transversal distributions of the intensity and polarization of light at double frequency (characterized by $M_2(x,y) = (|E_{2+}|^2 - |E_{2-}|^2)/(|E_{2+}|^2 + |E_{2-}|^2)$ and $\Psi_2(x,y) = 0.5\arg\{E_{2+}E_{2-}^*\}$, where $E_{2\pm} = E_{\perp x} \pm iE_{\perp y}$) may become more complicated.

Our investigations have shown that the parameters of the nonlinear medium practically do not affect the appearance and the behavior of the C-points in the center of the reflected second-harmonic beam. The C-point appears in the center of the beam if one of the conditions $a_x = ia_y$ (clockwise) or $a_x = -ia_y$ (counterclockwise rotation of the polarization in the C-point) and the polarization of $\mathbf{E}_\perp^{(NG)} \ne 0$ is different from the polarization in the C-point (the last condition is practically always true). Each of the equalities $a_x = ia_y$ or $a_x = -ia_y$ yields the equations set (two equations in each set: one for real and one for imaginary part of each of equalities). These equations sets were solved numerically for several combinations of the fixed values of the medium parameters. As a result for every value of the angle of incidence θ_1 we determined values of the ellipticity degree M_1 of the polarization ellipse and the angle of the orientation of the polarization ellipse Ψ_1 of the incident radiation corresponding to the appearance of the C-point in the center of the second-harmonic beam. This allows one to build dependencies $M_1(\theta_1)$ and $\Psi_1(\theta_1)$ for a given set of fixed medium parameters, such that if the polarization of the incident beam follows accordingly to these dependencies, the C-point remains in the center of the second-harmonic beam (actually we have two different pairs of $M_1(\theta_1)$ and $\Psi_1(\theta_1)$ for clockwise and counterclockwise rotation C-points).

Typical dependencies of $M_1(\theta_1)$ (solid line) and $\Psi_1(\theta_1)$ (dashed line) are shown at figure 1, plotted for the fixed values of medium parameters. Let us remark especially that for $\theta_1 = 0$ the value $\Psi_1(0)$ can be chosen arbitrarily due to the symmetry of the medium (it will be discussed later). It is convenient to count $\Psi_1(0) = \lim_{\theta_1 \to 0} \Psi_1(\theta_1)$ topological charge of the appearing C-point does not change when changing the angle of incidence with the simultaneous change of the polarization state of the incident beam ($M_1(\boldsymbol{\theta}_1)$ and $\Psi_1(\boldsymbol{\theta}_1)$). If the topological charge is positive, the type of the C-point can alternate from lemon to monstar and vice versa. For a given fixed value of $\boldsymbol{\theta}_1$ small variations of the polarization state of the incident radiation do not lead to the disappearance of the C-point, but slightly change its position in the signal beam cross-section, moving it from the center.

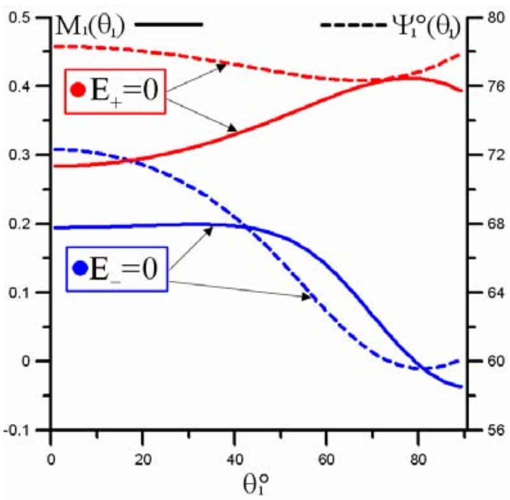

Figure 1. The dependencies of the polarization parameters of the incident radiation (the ellipticity degree M_1 (solid line) and the angle of orientation of the polarization ellipse Ψ_1 (dashed line)) on the angle of incidence θ_1 corresponding to the appearance of the C-point (with clockwise ($E_- = 0$) either counterclockwise ($E_+ = 0$) rotation) in the center of the second harmonic beam cross-section.

It is worth to notice that separate changes of M_1, Ψ_1 and $\boldsymbol{\theta}_1$ move the C-point in different directions. The C-point does not disappear in these cases due to the fact that the electric field $\mathbf{E}_{x,y}$ continuously depend on the parameters of the incident radiation so, that small variations of these parameters cannot result in abrupt changes of the light polarization distribution in the beam at double frequency. Right upper corner insets show polarization streamlines in the vicinity of the C-point in each case.

One should pay special attention to the case when the angle of incidence is small or equal to zero. Figure 2 shows the inhomogeneous polarization distribution at the double frequency beam for the same set of parameters as in fig. 1. Ellipses in different points of the pictures have the same shape as the light polarization ellipses in the same points of the beam cross-section. The dot at the edge of each of the ellipses indicates the position of the end of the electric field vector at fixed timing, i.e., it determines the angle $\Phi_2 = \arg\{E_{2+} + E_{2-}^*\}$ between the x-axis and the electric field vector. For all parts of this figure the sense of rotation of the electric field vector remains the same (clockwise) along the cross-section of the beam. The size of the ellipse (the sum of squared axes of the ellipse) is proportional to the intensity of light which is normalized at maximum intensity in each picture. Right upper corner insets show the direction of the polarization streamlines (the main axes of the ellipses are tangent to these lines in each point) in the vicinity of the C-point.

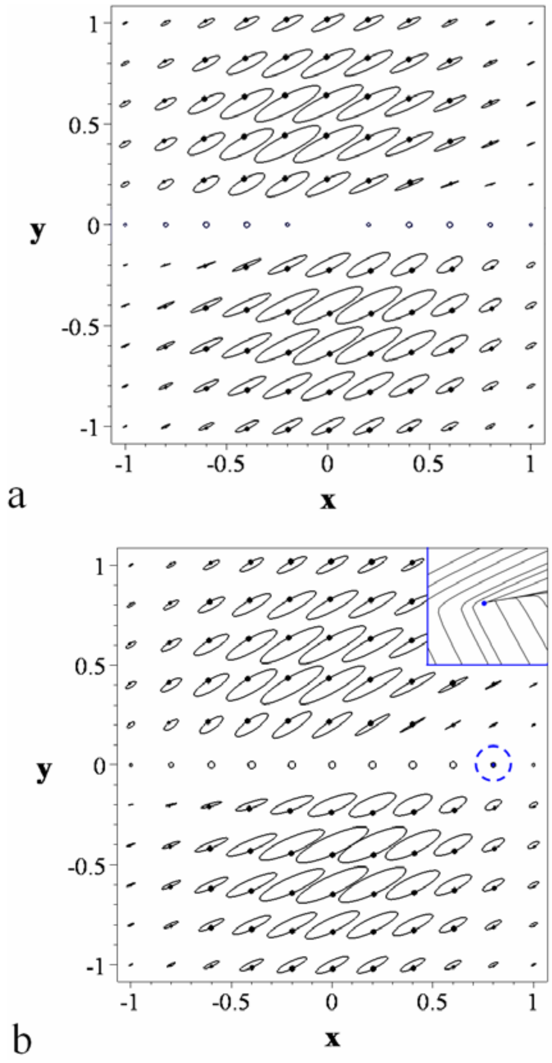

Figure 2. The polarization distribution in the reflected beam at double frequency: (a) normal incidence, the C-line is horizontal; (b) oblique incidence $\boldsymbol{\theta}_1 = 2°$, such that the axis of the incident beam lies in the same plane as the perpendicular to the surface and the C-line shown in (a); the C-line is transformed into the C-point.

If $\boldsymbol{\theta}_1 = 0$ (figure 2a), then the coefficients a_{3x} and a_{3y} become zero, and reflected double frequency beam attains additional symmetry: the polarization state of light in this beam depends only on the polar angle coordinate (and not on the polar radius). In other words the ellipticity degree and the angle of orientation of the polarization ellipse keep unchanged along the arbitrary straight line in the cross-section of the signal beam crossing the center of the beam (the intensity is zero in the center).

Thus, if $\theta_1 = 0°$ there cannot appear any point singularities in the cross-section of the signal beam but only L-lines and C-lines. At figure 2a plotted for $M_1 = 0.195$ and $\Psi_1 = 72.34°$ the C-line appears as a horizontal line. For any other value of Ψ_1 the polarization distribution and the C-line will be rotated by the angle $\Psi_1 - 72.34°$ around the center of the beam (z-axis) without changes in its structure due to the symmetry of the medium.

In the geometry of normal incidence the appearance of the C-lines is possible only when certain condition on the M_1 and the medium parameters is satisfied, which can be considered as the dependence of the ellipticity degree of the polarization of the incident radiation on the parameters of the medium. The position of the C-line with counterclockwise (clockwise) electric field rotation in the transversal section of the reflected signal beam is determined by the angle φ_{C-} (φ_{C+}) between the line and the main axis of the polarization ellipse of the incident radiation (since the normally reflected signal beam and normally incident fundamental beam cross-sections lie in the same plane).

This means that the choice of $\Psi_1 = -\varphi_{C+}$ or $\Psi_1 = -\varphi_{C-}$ makes the C-line to be oriented along the x-axis in the reflected beam. When passing across the C-line in the cross-section of the beam, the angle of orientation of the polarization ellipse suffers $\pi/2$-jump. We remark especially that both values of the ellipticity degree M_{1+} and M_{1-} exist for any real values of medium parameters, i.e. in the absence of absorption at frequencies ω and 2ω.

Now let us incline the incident beam for a small angle θ_1 from the vertical position in such a way, that the beam axis, the C-line (in the normally reflected signal beam) and the perpendicular to the surface remain in the same plane (see figure 3). In order to find the electric field distribution in the reflected beam we use formula (3). This formula was obtained assuming the plane of incidence being the $x_1 z_1$-plane in the coordinate system of the incident beam (xz-plane in the reflected beam, XZ-plane of the medium coordinate system). This means that in order to incline the incident beam as described above one needs to have a C-line oriented along the x-axis in the reflected signal beam. This is possible when the value of Ψ_1 for the case of normal incidence was chosen to be $\Psi_1 = -\varphi_{C-} = 72.34°$. Now let us remark that the values of the dependencies $M_1(\theta_1)$ and $\Psi_1(\theta_1)$ tend to values $M_{1\pm}$ and $-\varphi_{C\pm}$ correspondingly for $\theta_1 \to 0$. For example, the dependencies at figure 1 are built for the same parameters as the polarization distributions at figure 2. One can see that for the lower two curves at figure 1 corresponding to the zeros of the E_--component the following is true: $M_1|_{\theta_1 \to 0} = M_{1-} = 0.195$, $\Psi_1|_{\theta_1 \to 0} = -\varphi_{C-} = 72.34°$.

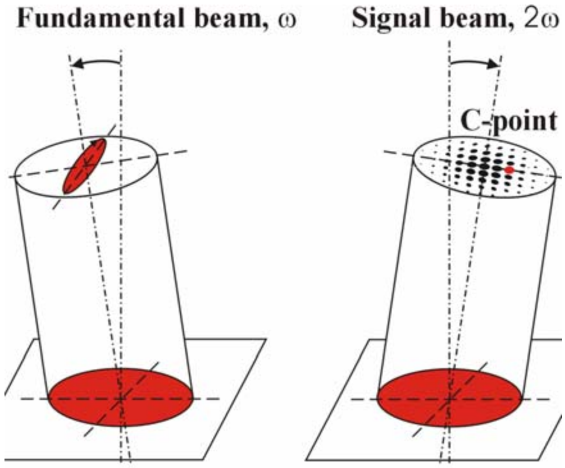

Figure 3. The scheme of transformation of the C-line into the C-point: oblique incident fundamental beam is shown at the left (the axis of the beam, the C-line (dashed line along the transversal section of the beam) in the normally reflected beam and the perpendicular to the surface remain in the same plane), and the reflected signal beam with the C-point, which is the result of the transformation of the C-line, is shown at the right.

After inclination of the incident beam for the small angle θ_1 one can see that C-point appears close to the center of the reflected second harmonic beam (figure 2 b, $\theta_1 = 2°$). This can be explained by the fact that for this small values of θ_1 "current" values of $M_1 = M_{1-}$ and $\Psi_1 = -\varphi_{C-}$ are still close though not equal to the $M_1(\theta_1)$ and $\Psi_1(\theta_1)$, because the dependencies at figure 1 have horizontal tangent lines in the point $\theta_1 = 0°$.

When increasing the angle of incidence the C-point moves away from the center along the direction which coincides with the direction of the C-line in the cross-section of normally reflected signal beam. For small θ_1 this can be qualitatively explained in the following way: small variation of the angle of incidence near the normal incidence ($\theta_1 = 0$) is equivalent to the variation of the transversal component of the wave vector in the Fourier representation of the light beam. Thus, we efficiently change the direction of the wave vector of the spatial Fourier component responsible for the appearance of the singularity in the beam. In this case the C-point moves away from the center in the direction which is opposite to the inclination direction, i.e., in the positive direction of the x-axis.

When inclining the incident beam in any other direction which does not coincide with the C-line (by appropriate choice of the Ψ_1 making the position of the C-line, for example, perpendicular to the x-axis, the value of Ψ_1 is too far from the $\Psi_1(\theta_1 = 0)$ and C-point does not appear in the reflected signal beam.

Conclusion

We have studied the conditions of the appearance of the C-points and their behaviour in the cross-section of the reflected beam at double frequency arising in case of incidence of the uniformly elliptically polarized Gaussian beam at fundamental frequency on the surface of the isotropic chiral medium with spatial dispersion of quadratic nonlinearity. The studies were performed in the assumption that the divergence angle $\theta_{div} \approx \lambda / w$ of the incident beam is small, and that $90° - \theta_1 > \theta_{div}$. It is established that for fixed values of the nonlinear medium parameters for every value of θ_1 there exists such a polarization state of the incident beam (with the ellipticity degree $\widetilde{M}_1(\theta_1)$ and the orientation angle of the main axis of the polarization ellipse $\widetilde{\Psi}_1(\theta_1)$) that there appears a C-point in the center of the second harmonic beam. Small variations of the ellipticity degree M_1 and the angle Ψ_1 of the incident beam near the values $\widetilde{M}_1(\theta_1)$ and $\widetilde{\Psi}_1(\theta_1)$ result in a shift of the C-point away from the center of the beam. Moreover, separate changes of $\widetilde{M}_1(\theta_1)$, $\widetilde{\Psi}_1(\theta_1)$ or θ_1 when the other two parameters are fixed move the C-point in different directions within the transversal section of the second harmonic beam.

When decreasing the angle of incidence of the fundamental beam to zero along with the according change of the polarization of this beam providing the presence of the C-point in the center of the second harmonic beam, then this C-point transforms into the C-line for $\theta_1 = 0$ at normal incidence. The angle of the orientation of this C-line in the cross-section of the second harmonic beam and the value of the ellipticity degree of the polarization ellipse of the fundamental beam, yielding the appearance of this line can be expressed by simple analytical formulae. It is noticeable that the polarization state corresponding to the ellipticity degree can be uniquely found for given real values of the medium parameters. Thus, choosing the $\widetilde{M}_1(0)$ and $\widetilde{\Psi}_1(0)$ as the initial polarization of the incident beam and then increasing the angle of incidence (inclining the beam) in such a way that the C-line, the incident beam axis and the perpendicular to the surface remain in the same plane, it is possible ot observe the transformation of the C-line to the C-point in the cross-section of the second harmonic beam. Further increase of the angle of incidence (up to grazing angles) with according change of the polarization of the incident beam (like in figure 1) will maintain the C-point in the center of the reflected second harmonic beam.

Therefore, for any real values of the medium parameters and for any values of the angle of incidence θ_1 (apart those which are close to $90°$, such that the angle of divergence of the incident beam becomes bigger than $90° - \theta_1$) there exists such a polarization state of the incident beam, that there appears a C-point with clockwise or counterclockwise rotating electric field vector in the center of the reflected second harmonic beam.

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