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Singularities of the second-harmonic light field polarisation arising upon reflection of normally incident elliptically polarised Gaussian beam from the surface of an isotropic chiral medium

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Abstract. **We have analysed the conditions for the appearance of polarisation singularities in the second-harmonic beam cross section arising in the case of reflection of a uniformly elliptically polarised Gaussian beam at the fundamental frequency from the surface of an isotropic gyrotropic medium. It is shown that there are elliptical polarisation states of the incident light at which the cross section of the second-harmonic reflected beam contains either one or two C lines and either two, or one, or none L lines [the loci of the points where the propagating radiation is circularly (C) or linearly (L) polarised].The formulas determining the conditions for the occurrence of L and C lines and specifying their orientation in the plane of the cross-section of the second-harmonic beam are obtained.**

Keywords: *polarisation singularities, second-harmonic generation, chirality, nonlinear-response nonlocality.*

The laser-beam second-harmonic generation (SHG) was discovered 50 years ago. Nevertheless, it has been actively investigated, applied, and improved in different schemes. For example, SHG is currently used to study the local and nonlocal responses of nanoparticles [1] and microstructured materials [2] and to analyse the radiation scattering from nanoscale surface inhomogeneities [3]. There have also been attempts to apply SHG for studying the two-photon resonances and vibrational spectra of various molecules [4–6].

One of the widest spread methods for studying nonlinear optically active media and thin films is based on the SHG from their surface. The conditions for second-harmonic generation and the technique for detecting the effects caused by the chirality of molecules are well known and described in the literature [7–12]. A relatively long time ago some theoretical studies were carried out within the plane-wave approximation, where the influence of the spatial dispersion of the nonlinear optical response of a chiral medium [7–10] and the surface inhomogeneity of its optical properties [13] were taken into account in different ways. In some experiments there were attempts to separate the contributions from the surface and volume of the material to the second-harmonic signal [14] and select the second-harmonic component that is due to the chirality of the medium [15, 16]. The authors of [11, 12] were the first to take into account the boundedness of the fundamental beam when calculating the intensity and polarisation parameters of the second-harmonic signal from the surface of a medium with a spatial dispersion of quadratic nonlinearity. Formulas for the transverse spatial Fourier transform of the electric field of the second-harmonic reflected wave in the far-field diffraction zone were obtained. However, these studies were focused on the possibility of second-harmonic generation and calculation of the energy characteristics of the reflected second-harmonic radiation (recall that within the plane-wave approximation SHG cannot occur when a fundamental wave is normally incident on an optically active medium). In these studies the surface inhomogeneity of the optical properties of a nonlinear medium was taken into account using modified boundary conditions for the electric and magnetic fields. The correctness of this approach was substantiated in [17, 18].

Thereafter, the formulas describing the spatial distribution of polarisation of the propagating second harmonic were found and analysed in [19]. Despite the results of the studies, indicative of the occurrence of spatially nonuniform polarisation distributions in the second-harmonic beam, no studies were aimed at revealing the possibilities and conditions for the formation of singularities of light-field polarisation in this beam (points or lines in the propagating beam cross section, where the intensity of one of its two orthogonal polarised components becomes zero).

The conditions for the occurrence of polarisation singularities and the dynamics of their propagation in space were multiply theoretically and experimentally studied in linear optics. A terminology (which is actively used nowadays) was developed in one of the first studies [20]; according to it, the loci of the points where the propagating radiation has a circular (linear) polarisation are referred to as C lines (L surfaces). In the beam cross-section plane they are transformed into C points (L lines). In contrast to the conventional optical vortices (or screw phase dislocations) with zero scalar-field intensity (they are generally studied within the approximation of constant polarisation of the propagating radiation), the C points (where the orientation of the polarisation ellipse of the electric field of electromagnetic wave is not determined) can be referred to as 'component' optical vortices. The morphological distributions of the polarisation ellipses in their vicinities can be of three types: 'star', 'lemon', and 'monstar'.

The conditions for the formation of polarisation singularities and the dynamics of their development in various problems of linear optics have been investigated later [20–31]. We should also note the highly efficient experimental methods for detecting light beams with phase and polarisation singularities [26–31]. The polarisation singularities are stable

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objects in the propagating light beam, and their formation, annihilation, and interaction follow strictly specified scenarios.

This work was stimulated by the absence of studies on the formation of polarisation singularities in the signal beam in the case of second-harmonic or sum-frequency generation from the surface of an isotropic chiral medium and by the small number of studies on the formation of polarisation singularities in the nonlinear optics applications. We considered the polarisation singularities arising in the cross section of the double-frequency beam that is formed during SHG by a uniformly polarised fundamental Gaussian beam normally incident on the surface of an isotropic chiral medium. Specifically under the conditions of normal incidence the nonuniformity of the polarisation distribution in the signal beam is most pronounced [12]. In this case, the polarisation distribution in the reflected beam is cylindrically symmetric [19], i.e., the polarisation state is the same along any straight line drawn in the cross-section plane of the beam through its centre. This condition forbids point singularities (C points) in the beam but allows for L and C lines intersecting in the beam centre, where the intensity is zero. In this paper, we report the results of studying these objects.

Let an elliptically polarised monochromatic Gaussian beam be normally incident on the flat surface of an isotropic gyrotropic medium from vacuum. The electric-field strength in this beam is given by the formula

$$\boldsymbol{E}(x,y,z) = \left[\boldsymbol{e} + \frac{\mathrm{i}\boldsymbol{e}_z(\boldsymbol{e}\nabla)}{k}\right]\frac{E_0}{\beta(z)}\exp\left(-\frac{x^2+y^2}{w^2\beta(z)} - \mathrm{i}\omega t + \mathrm{i}kz\right)$$

$$= E_\perp \boldsymbol{e} + E_z \boldsymbol{e}_z. \qquad (1)$$

Here, $\boldsymbol{e}_z$ is the unit vector parallel to the z axis; E_0 is the amplitude; ω is the frequency; w is the incident-beam half-width; $k = \omega/c$ is the wave-vector magnitude; and $\beta(z) = 1 + 2\mathrm{i}z/(kw^2)$. Expression (1) contains the longitudinal component $E_z\boldsymbol{e}_z$ and the transverse component $E_\perp\boldsymbol{e} = [E_+(\boldsymbol{e}_x - \mathrm{i}\boldsymbol{e}_y) + E_-(\boldsymbol{e}_x + \mathrm{i}\boldsymbol{e}_y)]/\sqrt{2}$, where $E_\pm = E_x \pm \mathrm{i}E_y$ are the circularly polarised field components and $\boldsymbol{e}_x$ and $\boldsymbol{e}_y$ are the unit vectors parallel to the x and y axes, respectively. It is convenient to characterise the incident light beam by the normalised intensity $I(x,y) = (|E_+|^2 + |E_-|^2)/2$, which depends on the transverse coordinates, and two constants: the ellipticity of the polarisation ellipse, $M_0 = (|E_+|^2 - |E_-|^2)/(|E_+|^2 + |E_-|^2)$, and the inclination angle of its major axis, $\Psi_0 = 0.5\arg\{E_+E_-^*\}$. The parameter M_0 is generally varied from –1 (left-handed circular polarisation) to 1 (right-handed circular polarisation), with a passage through zero (linear polarisation). The isotropic gyrotropic medium belongs to the limiting group $\infty\infty$, and its surface belongs to the group ∞. Therefore, the angle Ψ_0 can be equated to zero without losing generality (actually, the x-axis direction is specified by the direction of the major axis of the incident-radiation polarisation ellipse). In this case, the unit complex polarisation vector $\boldsymbol{e} = 0.5[(1 - M_0)^{1/2}(\boldsymbol{e}_x + \mathrm{i}\boldsymbol{e}_y) + (1 + M_0)^{1/2}(\boldsymbol{e}_x - \mathrm{i}\boldsymbol{e}_y)$. Note that expression (1) satisfies the Maxwell equation $\mathrm{div}\boldsymbol{E} = 0$ within the first-order approximation for the beam divergence angle, which will be considered as small below.

The distribution of the electric field $\boldsymbol{E}_{\mathrm{SH}} = E_{\mathrm{SH}+}\boldsymbol{e}_- + E_{\mathrm{SH}-}\boldsymbol{e}_+$ in the beam is given by the formula

$$E_{\mathrm{SH}\pm}(r,\varphi,z) = \frac{32\pi i E_0^2 r}{w^2\omega(1+n_\omega)^2(1+n_{2\omega})[\beta^*(z)]^2}$$

$$\times\{-C_0\sqrt{1-M_0^2}\exp(\pm\mathrm{i}\varphi) + [\sqrt{1+M_0}\exp(-\mathrm{i}\varphi)$$

$$+\sqrt{1-M_0}\exp(\mathrm{i}\varphi)]\sqrt{1\pm M_0}\,C_\pm\}$$

$$\times\exp[-2\mathrm{i}\omega t - \mathrm{i}2\omega z/c - 2r^2/(w^2\beta^*(z))], \qquad (2)$$

which was derived previously in [19]. Here, $r = (x^2 + y^2)^{1/2}$ and $\varphi = \arctan(y/x)$ are, respectively, the polar radius and angle in the cylindrical coordinate system; $C_0 = n_{2\omega}b_1 + \mathrm{i}\gamma_0/n_{2\omega}$; $C_\pm = n_\omega(b_3 \mp \mathrm{i}b_5)$; n_ω and $n_{2\omega}$ are the linear refractive indices of the medium at frequencies ω and 2ω; $b_1 = \kappa^{(2)}_{zxx} = \kappa^{(2)}_{zyy}$; $b_3 = \kappa^{(2)}_{yyz} = \kappa^{(2)}_{xxz} = \kappa^{(2)}_{yzy} = \kappa^{(2)}_{xzx}$; $b_5 = \kappa^{(2)}_{xyz} = -\kappa^{(2)}_{yxz} = \kappa^{(2)}_{xzy} = -\kappa^{(2)}_{yzx}$; The tensors $\hat{\gamma}^{(2)}(2\omega;\omega,\omega)$ and $\hat{\kappa}^{(2)}(2\omega;\omega,\omega)$ describe, respectively, the spatial dispersion of the quadratic nonlinearity of the chiral medium and the field-quadratic response of its surface within the approach developed in [17, 18]. The numerical values of the parameters characterising the nonlinearity of isotropic chiral liquids and a review of the experimental studies of these parameters can be found in [19, 32].

The condition for the occurrence of the L line $\varphi = \varphi_{\mathrm{L}}$, which makes the angle φ_{L} with the x axis, in the cross section of the second-harmonic beam is

$$|E_{\mathrm{SH}+}(r,\varphi=\varphi_{\mathrm{L}},z)| = |E_{\mathrm{SH}-}(r,\varphi=\varphi_{\mathrm{L}},z)|. \qquad (3)$$

At the points belonging to this line the ellipticity $M_{\mathrm{SH}}(r, \varphi, z) = (|E_{\mathrm{SH}+}|^2 - |E_{\mathrm{SH}-}|^2)/(|E_{\mathrm{SH}+}|^2 + |E_{\mathrm{SH}-}|^2)$ of the second-harmonic polarisation ellipse becomes zero. It was shown in [19] that, at certain values of the nonlinear-medium parameters and the incident light ellipticity, Eqn (3) may have two roots,

$$\varphi_{\mathrm{L1,L2}} = \arctan[(-B \pm \sqrt{B^2 - AC})/A], \qquad (4)$$

if the values

$$A = (1 - \sqrt{1-M_0^2})[M_0 n_\omega(b_3^2 + b_5^2) + b_5\gamma_0/n_{2\omega}]$$

$$- M_0[M_0 b_5\gamma_0/n_{2\omega} + \sqrt{1-M_0^2}\,n_{2\omega}b_1b_3],$$

$$B = \sqrt{1-M_0^2}\,(b_3\gamma_0/n_{2\omega} + n_{2\omega}b_1b_5M_0),$$

$$C = M_0 n_\omega(b_3^2 + b_5^2) + (1 - M_0^2)b_5\gamma_0/n_{2\omega}$$

which enter into (4), satisfy the condition $B^2 - AC > 0$. In this case, the cross section plane of the second-harmonic beam is divided by two L lines ($\varphi = \varphi_{\mathrm{L1}}$ and $\varphi = \varphi_{\mathrm{L2}}$), which are intersected at the point (0, 0, z) into four sectors, so that the polarisation rotation direction changes every time when passing through the L line. At $B^2 - AC = 0$ we have $\varphi_{\mathrm{L1}} = \varphi_{\mathrm{L2}}$, and there is one L line in the beam and the polarisation rotation is the same everywhere. If $B^2 - AC < 0$, Eqn (4) has no solutions. Polarisation singularities of the L type do not arise in this case.

The conditions for the occurrence of C line ($\varphi = \varphi_{\mathrm{C}+}$) ($\varphi = \varphi_{\mathrm{C}-}$) with left-handed (right-handed) rotation of the electric-field vector, where $M_{\mathrm{SH}}(\varphi_{\mathrm{C}+}) = -1$ ($M_{\mathrm{SH}}(\varphi_{\mathrm{C}-}) = 1$), can be written, respectively, as

$$E_{\mathrm{SH}+}(r,\varphi=\varphi_{\mathrm{C}+},z) = 0, \qquad (5)$$

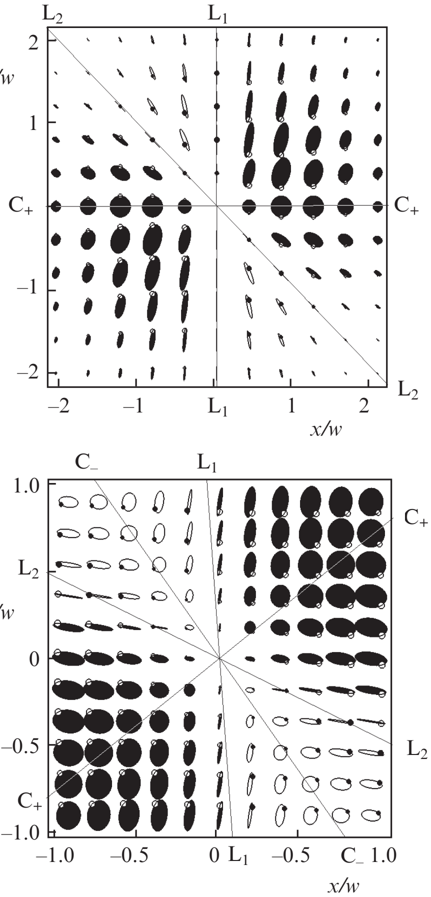

Figure 1. Nonuniform distribution of light polarisation in the second-harmonic reflected beam at (a) $M_0 = 0$, $n_\omega = 1.33$, $n_{2\omega} = 1.35$, $b_3/b_1 = 0.508$, $b_5 = b_3$, $\gamma_0/b_1 = -1.823$ (there is one C line and two L lines) and (b) $M_0 = -0.27$, $n_\omega = 1.32$, $n_{2\omega} = 1.34$, $b_3/b_1 = 1.1$, $b_5 = 0.322$, $\gamma_0/b_1 = -2.1$ (there are two C lines and two L lines). The ends of the C and L lines are marked by symbols L_1, L_2, C_+ and C_-.

$$E_{SH-}(r, \varphi = \varphi_{C-}, z) = 0. \tag{6}$$

Actually, each of Eqns (5) and (6) is a system of two equations, because the quantities $E_{SH\pm}(r, \varphi, z)$ are complex. Therefore, their solutions $\varphi = \varphi_{C+}$ and $\varphi = \varphi_{C-}$, exist only under some limitation on the parameters M_0, b_1, b_3, b_5 and γ_0; it is reasonable to present this limitation as a dependence of the polarisation of incident radiation on the parameters of nonlinear medium. At

$$M_0 = M_{0\pm}(b_1, b_3, b_5, \gamma_0) = \pm\frac{G_\pm - G_1}{G_\pm + G_1}, \tag{7}$$

where $G_1 = n_\omega^2(b_3^2 + b_5^2)$ and $G_\pm = (\gamma_0/n_{2\omega} \pm n_\omega b_5)^2 + (n_\omega b_3 - n_{2\omega} b_1)^2$, the solutions to (5) and (6) take, respectively, the form

$$\varphi = \varphi_{C\pm} = \arctan\frac{n_\omega b_3\sqrt{G_\pm} + (-n_{2\omega}b_1 + n_\omega b_3)\sqrt{G_1}}{n_\omega b_5\sqrt{G_\pm} - (\pm\gamma_0/n_{2\omega} + n_\omega b_5)\sqrt{G_1}}. \tag{8}$$

Having passed through the line $\varphi = \varphi_{C+}$ or $\varphi = \varphi_{C-}$ in the beam cross-section plane, the angle of rotation of the major axis of the polarisation ellipse changes stepwise by $\pi/2$. Note that both values of the ellipticity $M_{0\pm}$ exist at any real values of b_1, b_3, b_5 and γ_0 (i.e., in the absence of absorption at the frequencies ω and 2ω). Only when all components of the tensors $\hat{\gamma}^{(2)}(2\omega;\omega,\omega)$ and $\hat{\kappa}^{(2)}(2\omega;\omega,\omega)$ are zero (the double-frequency signal is absent), $G_\pm + G_1 = 0$.

For example, Fig. 1 shows the polarisation distributions in the cross section of the second-harmonic beam. The ellipses plotted in the different areas of the figure, with centres at the coordinates x_0, y_0, are similar to the light polarisation ellipses at the points with the same coordinates in the beam cross section. The angle between the major axis of the ellipse centred at the point (x_0, y_0) and the x axis in Fig.1 coincides with the inclination angle of the major axis of the second-harmonic polarisation ellipse: $\Psi_{SH}(x_0,y_0) = 0.5\times \arg\{E_{SH+}E^*_{SH-}\}$. The point at the edge of each of them specifies the electric field direction at a fixed instant (i.e., determines the angle $\Phi_{SH} = \arg\{E_{SH+} + E^*_{SH-}\}$). The open and closed ellipses indicate that the electric field at the point (x_0, y_0) is rotated clockwise and counterclockwise, respectively.

Figure 1a shows the polarisation distribution in the second-harmonic beam cross section at the nonlinear-medium parameters allowing for the presence of one C line and two L lines in this cross section. The C line coincides with the x axis. The ellipse on this line transforms into a circle. When passing through the C line, the rotation angle of the major axis of the polarisation ellipse changes stepwise by $\pi/2$. Two L lines are intersected at the beam centre. One of them is vertical, while the other makes an angle of 45° with it and passes through the second and fourth quadrants of the xy coordinate system. Figure 1b was plotted at the parameters of the incident radiation and nonlinear chiral medium that allow for the occurrence of two C lines and two L lines in the cross section plane of the second-harmonic beam. All these lines pass through the beam centre. One of the C lines passes through the first and third quadrants, while the other passes through the second and fourth quadrants. One of the L lines is almost vertical, while the other passes through the second and fourth quadrants.

Note a number of peculiar cases, which are implemented at specific ratios of the parameters b_1, b_3, b_5 and γ_0. If $n_{2\omega}b_1 = n_\omega b_3$ and condition (7) is satisfied, the reflected second-harmonic beam is left- (at $\gamma_0 = -n_\omega n_{2\omega} b_5$) and right-hand ($\gamma_0 = n_\omega n_{2\omega} b_5$) circularly polarised at all points of the cross section. If there is no spatial dispersion ($\gamma_0 = 0$) but condition (7) is valid and at least one of the equalities $b_1 = 0$ or $n_{2\omega}b_1 = 2n_\omega b_3$ is satisfied, the second-harmonic beam is linearly polarised at all points of the cross-section plane. In this case, this plane contains a straight line passing through the beam centre; the second-harmonic field intensity becomes zero at the points of this line, because the orientations of two C lines coincide. Finally, if the properties of the medium satisfy the equalities $b_3 = b_5 = 0$, the second-harmonic beam is radially polarised (see [19]). In this case, condition (7) is transformed into $M_0 = \pm 1$, and the second-harmonic signal disappears.

Analytical expressions (4), (7), and (8) are of practical interest for nonlinear laser spectroscopy. Indeed, for linearly polarised incident radiation, the polar angles (4), which determine the orientation of the L lines in the cross-section plane of the second-harmonic beam, become equal to $\pi/2$ and $-\arctan(b_5/b_3)$. If the integral beam power in the case of circularly polarised pumping is known, one can find the b_3 and b_5 values (see [19] for details). If the n_ω and $n_{2\omega}$ values are known, relations (4), (7), and (8) can be used to determine γ_0 and b_1.

Thus, under normal incidence of a uniformly elliptically polarised fundamental Gaussian beam on the surface of an isotropic gyrotropic medium, polarisation singularities may occur in the cross section of the reflected second-harmonic beam. Any real values of the parameters describing the quadratic optical response of a chiral medium and its surface allow for such states of elliptical polarisation of the incident light that the cross section of reflected second-harmonic beam would contain one or two C lines. Exceptions are few cases where the parameters of the medium satisfy exactly the peculiar relations between them. The cross section of the second-harmonic beam may also contain two, one, or none L lines. The analytical expressions describing the conditions for the occurrence of L and C lines and the formulas determining their orientation can be used to find the components of the tensors characterising the field-quadratic nonlocal optical response of the medium and the local response of its surface. The case of oblique incidence, which is much more complex, is of peculiar interest. Determination of the conditions for the occurrence of polarisation singularities in this case is beyond the scope of this study.

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