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Generation of reflected second-harmonic light beam with inhomogeneous transversal distribution of polarization from the surface of chiral medium by normally incident Gaussian beam

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Abstract

A principle possibility of second harmonic generation (SHG) from the surface of a chiral medium by normally incident focused fundamental beam has been shown earlier, and the key features of this phenomenon (forbidden in a planewave approximation) have been outlined in [N.I. Koroteev, V.A. Makarov, S.N. Volkov, Laser Phys. 8 (1998) 532–535]. In our work we have obtained analytical expressions, which describe the distributions of intensity and polarization in the cross-section of a second-harmonic (SH) light beam. It is found that the polarization state drastically changes along the cross-section of the signal light beam. The polarization effects concerning the transversal inhomogeneous polarization distribution were studied in detail. It is shown that the measurement of the polarization state in certain areas of the SH beam cross-section gives us quantitative information directly about the medium material constants.

Keywords: Second harmonic generation; Elliptic polarization; Spatial dispersion; Gyrotropy; Chirality; Surface; Inhomogeneous polarization distribution

1. Introduction

Surface second harmonic generation (SHG) is very often used as a tool for the investigation of complex organic molecules, which are located in surface layers or in thin films. The conditions of the appearance of second harmonic (SH) signal and the methods of detection of the response provided by the chiral properties of the molecules are well known and described in the literature [2–7]. Various schemes of SHG and ellipsometry of light at double frequency are continuously being improved. These are used for various applications such as surface diagnostic and spectroscopy studies [8–12], and in the control of film growth [13]. Modern spectroscopic studies analyze the rotation of the polarization plane of SH, the difference between the intensities of its linearly (or circularly) polarized components [14–16], and develop and improve the methods of separation of the signal at double frequency provided by the medium chirality [17].

In theoretical studies carried out within the framework of planewave approximation the authors made attempts by different ways to take into account the spatial dispersion of a nonlinear optical response of a chiral medium [18–21] as well as the surface layer inhomogeneity of the medium optical properties [22], and also to find ways of experimental separation of bulk and surface contributions in the SH signal [23]. The interconnection between the orientation of the molecules at the surface and the characteristics of the generated SH was also studied [24].

It is necessary to remark the importance of considering the spatial finiteness of light beams in their reflection from the border between two media. Already a long time ago it has been shown, that the effects having the first and the second order on the incident beam divergence angle give small (but essential) corrections to the laws of geometric optics

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for the reflection and refraction of light beams (both for 2D-beams [25] and 3D-beams [26]; see also Refs. 1–7 in [25] and experimental work [27]). Also these effects are responsible for the changes of the intensity and polarization distributions in the reflected and refracted beam cross-sections. Intense research in this area is also continuing presently [28–30]. In particular, these effects play a special role when the reflected signal is absent within the planewave approximation (when the incident beam is polarized in the plane of incidence and the incidence angle is equal to the Brewster angle).

For the first time the authors of [1,31] have taken into account the spatial finiteness of fundamental and signal light beams in SHG from the surface of a nonlinear medium with a spatial dispersion of quadratic nonlinearity in their studies of intensity and polarization of the signal wave at double frequency. In [31] the oblique incidence of two-dimensional Gaussian beam (slit beam) has been considered, and the normal incidence of three-dimensional Gaussian beam has been studied in [1]. The optical inhomogeneity of the surface layer of a nonlinear medium has been taken into account by means of modified border conditions for electromagnetic fields. The substantiation of reasonableness of such an approach has been presented in [32,33]. It is necessary to remark that in the case of the normal incidence of a fundamental beam, SHG is forbidden within the planewave approximation. That is why in [1] the main attention had been paid to the question of possibility of SHG and calculation of characteristics of its intensity and power, using the formula obtained for the spatial Fourier-image of the reflected wave on double frequency in the far field zone. In general case this Fourier-image depends on the nonlocal optical quadratic susceptibility $\hat{\gamma}^{(2)}$ of the medium (because local susceptibility $\hat{\chi}^{(2)}$ of the medium bulk becomes zero in the case of SHG (unlike in sum-frequency generation)) and the surface optical quadratic susceptibility $\hat{\kappa}^{(2)}$. Tensors $\hat{\gamma}^{(2)}$ and $\hat{\kappa}^{(2)}$ describe the spatial dispersion of quadratic optical response of the medium and the nonlinearity of its surface correspondingly. The spatial transversal distribution of polarization of SH radiation has not been studied neither in this work nor later. The only exception is the illustration for the simplest case of linear polarization of the incident wave, which is considered in [1].

The transversal polarization distribution in the signal beam in the three-wave mixing processes can be very specific and deserves to be explored in detail. This is already confirmed by the results of [34], where the investigation of the transversal distribution of polarization in a sum-frequency beam has been performed for the process of sum-frequency generation in a bulk of isotropic gyrotropic medium by two collinear elliptically polarized Gaussian beams. It has been shown [34] that all the parameters of light field, namely, the intensity $I(r,\varphi,z)$, the ellipticity degree $M(r,\varphi,z)$ and the angle of rotation $\psi(r,\varphi,z)$ of the main axis of the polarization ellipse, the angle $\Phi(r,\varphi,z)$ of orientation of the electric field vector at fixed timing (indicating the phase difference along the beam cross-section) essentially change in the transversal dimension of the signal beam, depending on the polar angle coordinate φ. The appearance of the last parameter $\Phi(\mathbf{r},z)$ for spatially finite elliptically polarized light beams is caused by the inhomogeneity of oscillation phase distribution for the electric field vector, which is not the case, of course, for the plane wave.

The other reason promoting these studies of inhomogeneous polarization distributions is the possibility of their spectroscopic applications. It is generally recognized that the values of the components of material tensors characterizing the quadratic optical nonlinearity of the surface and spatial dispersion of optical nonlinearity of medium bulk are more easily obtained if one is analyzing the dependencies of the SH polarization on the parameters of the polarization of the incident wave and on the nonlinear medium constants rather than the dependence of the SH intensity on the same quantities [10, 15–17]. But even precise analysis of the polarization dependence performed in [9] for an achiral medium can give values of relation of certain components only in the absence of spatial dispersion of bulk nonlinearity. The measurements of the relative intensities of circularly or linearly polarized components of the SH signal in a number of cases lead to a complex system of nonlinear algebraic equations, which does not always allow the convenient extraction of the information about the components of $\hat{\gamma}^{(2)}$ and $\hat{\kappa}^{(2)}$.

The analysis of the transversal inhomogeneous polarization distributions and their dependence on the parameters of incident light and the nonlinear medium constants in terms of $M(\mathbf{r})$ and $\psi(\mathbf{r})$ (and, possibly, even $\Phi(\mathbf{r})$) provides an alternative and effective method of extraction of the information about the nonlinear susceptibilities of the bulk and the surface of the medium.

In this work we present the results of the studies of the formation of inhomogeneously polarized light beam at double frequency generated in reflection from the surface of a chiral medium in the case of a normally incident focused fundamental elliptically polarized light beam with a Gaussian intensity profile. The graphics represented by means of the analytical formulas obtained in this work illustrate the drastic changes of the polarization state along the cross-section of the signal beam.

2. Analytic solution of the problem

Let us consider (as in [1]) that an elliptically polarized broad Gaussian beam (with effective waist size $w >> \lambda$, where λ is the light wavelength) normally falls on the surface of the medium ($z = 0$)

$$\mathbf{E}_1(\mathbf{r}, z = 0) = \mathbf{e}E_0 \exp\{-\mathrm{i}\omega t - r^2/w^2\}. \quad (1)$$

Here E_0 is the scalar amplitude of the light field, $\mathbf{e}$ is the unit complex vector defining the polarization state of the incident radiation, in general case, $|\mathbf{e}|^2 = 1$. Its specific form depends on the orthogonal basis, which is used in a given task. We assume that the waist of the beam is located at the surface of the medium. The phase and the polarization of the incident beam are uniformly distributed in its cross-section.

As the source for our further considerations we use the formula for the transversal component of the Fourier-image of the reflected light field at double frequency found in [1] (if the incident wave is given as (1))

$$\tilde{\mathbf{E}}_{2\omega}(\mathbf{k}_{2\omega\perp}, z=0) = \frac{w^2E_0^2}{\omega(1+n_1)^2(1+n_{2\omega})}\{-(b_1n_{2\omega}+\mathrm{i}\gamma_0/n_{2\omega})(\mathbf{e}\cdot\mathbf{e})\mathbf{k}_{2\omega\perp} + 2n_1(\mathbf{k}_{2\omega\perp}\cdot\mathbf{e})[b_3\mathbf{e}-b_5[\mathbf{e}_z\times\mathbf{e}]]\}\exp\{-2\mathrm{i}\omega t - w^2k^2_{2\omega\perp}/8\}, \tag{2}$$

where $\mathbf{k}_{2\omega\perp}$ is the transversal part (lying in the xy-plane) of the wave vector $\mathbf{k}_{2\omega}$ of the SH, n_1 and $n_{2\omega}$ are the refraction indexes at frequencies ω and 2ω, respectively, $b_1=\kappa^{(2)}_{zxx}$, $b_3=\kappa^{(2)}_{xxz}$ and $b_5=\kappa^{(2)}_{xyz}$ are three of the four independent components of 3rd rank tensor $\hat{\kappa}^{(2)}$, which characterizes the quadratic optical response of the surface with symmetry ∞. This tensor has permutation symmetry in its last two indexes as a consequence of pump wave frequency degeneration. $\gamma_0=\omega\gamma^{(2)}_{xxyy}$ is proportional to one of three independent components of the 4th rank tensor $\hat{\gamma}^{(2)}$, which is responsible for the linear spatial dispersion of the quadratic optical bulk susceptibility of chiral medium (symmetry $\infty\infty$) [1,31–33].

In order to obtain the spatial distribution of light field in the reflected beam at double frequency, it is sufficient to a perform reverse Fourier-transform of (2). Taking into account the dependence of the SH light field on the propagation coordinate z within the framework of parabolic approximation, this spatial distribution is given as following:

$$E_{2\omega}(r,z) = \int\int \tilde{E}_{2\omega}(k_{2\omega\perp}, z=0)\exp\{\mathrm{i}k_{2\omega\perp}r - \mathrm{i}k_{2\omega}z - \mathrm{i}zk^2_{2\omega\perp}/2k_{2\omega}\}\mathrm{d}k_{2\omega\perp} \tag{3}$$

Constituting (2) in (3) and carrying out double integration, we obtain

$$\mathbf{E}_{2\omega}(\mathbf{r},z) = \frac{1}{\beta^2(z)}\frac{32\pi\mathrm{i}E_0^2}{w^2\omega(1+n_1)^2(1+n_{2\omega})} \times\{-(b_1n_{2\omega}+\mathrm{i}\gamma_0/n_{2\omega})(\mathbf{e}\cdot\mathbf{e})\mathbf{r} + 2n_1(\mathbf{r}\cdot\mathbf{e})[b_3\mathbf{e}-b_5[\mathbf{e}_z\times\mathbf{e}]]\} \times\exp\{-2\mathrm{i}\omega t - \mathrm{i}k_{2\omega}z - 2r^2/w^2\beta(z)\}, \tag{4}$$

where $\beta(z) = (1-4\mathrm{i}z/k_{2\omega}w^2)$.

In principle, formula (4) describes the electric field transversal distribution in the SH beam for both absorptive and non-absorptive media (for example, in far-off-resonance and in near-resonance cases). But further, for simplicity, we consider only non-absorptive media with real values of n_1, $n_{2\omega}$, b_1, b_3, b_5 and γ_0, though results of our work show that the same studies can be performed as well for absorptive media with complex material constants.

It is convenient to describe the SH light field considered in this work by the normalized intensity $I=(|E_+|^2+|E_-|^2)/2$, the ellipticity degree $M=(|E_+|^2-|E_-|^2)/(|E_+|^2+|E_-|^2)$ and the rotation angle $\Psi = 0.5\arg(E_+E_-^*)$ of the polarization ellipse, and also by the angle $\Phi=\arg(E_+ + E_-^*)$ between the electric field vector and the axis x of the coordinate system at fixed timing $t=k_{2\omega}z/_{2\omega}$. Of course, consideration of this parameter makes sense only if we are considering not a plane wave, but finite light beams. The expressions for these four parameters defined above contain $E_\pm = E_x \pm \mathrm{i}E_y$, which are the complex circularly polarized amplitudes of the light wave. Remarkably, $-1\leqslant M\leqslant 1$. $M=\pm1$ corresponds to the circular polarization states (with left or right rotation, depending on the sign of M), and $M=0$ corresponds to the linear polarization. With these definitions of the ellipticity degree and the angle of rotation of the polarization ellipse it is reasonable to define the polarization vector of the fundamental wave as the following: $\mathbf{e}=\sqrt{(1+M_0)/2}\cdot e^{\mathrm{i}\Psi_0}\mathbf{e}_- + \sqrt{(1-M_0)/2}\cdot e^{-\mathrm{i}\Psi_0}\mathbf{e}_+$, where $\mathbf{e}_\pm = (\mathbf{e_x}\pm\mathrm{i}\mathbf{e_y})/\sqrt{2}$, where $\mathbf{e}_{x,y}$ are the unit vectors in x- and y-axes positive directions. In this case M_0 is the ellitpicity degree of the polarization ellipse of the incident wave (correspondingly to our definition) and can be varied in the range of $-1\leqslant M_0\leqslant 1$. Since the medium is symmetric with respect to any rotation around the axis of the incident beam, the angle of rotation ψ_0 of the polarization ellipse of the incident wave can be taken equal to 0. At that, the polarization vector of the incident radiation will look as $\mathbf{e}=\sqrt{(1+M_0)/2}\,\mathbf{e}_- + \sqrt{(1-M_0)/2}\,\mathbf{e}_+$.

By using (4) it is not difficult to obtain the following cumbersome expressions for normalized intensity, ellipticity degree, angle Φ in the cylindrical coordinate system with coordinates r, φ and z

$$I(r,\varphi,z) = 0.5\,|D(r,z)|^2[4n_1^2(b_3^2+b_5^2) + (1-M_0^2) \times(2(n_{2\omega}^2b_1^2+\gamma_0^2/n_{2\omega}^2) - 4n_1n_{2\omega}b_1b_3) + \sqrt{1-M_0^2}\,\{[4n_1^2(b_3^2+b_5^2) - 4n_1n_{2\omega}b_1b_3 + 4n_1b_5M_0\gamma_0/n_{2\omega}]\cos 2\varphi + [4n_1n_{2\omega}b_1b_5 + 4n_1b_3M_0\gamma_0/n_{2\omega}]\sin 2\varphi\}] \tag{5}$$

$$M(\varphi) = [M_0n_1^2(b_3^2+b_5^2) + (1-M_0^2)n_1b_5\gamma_0/n_{2\omega} + \sqrt{1-M_0^2}\,\{[M_0(n_1^2(b_3^2+b_5^2) - n_1n_{2\omega}b_1b_3) + n_1b_5\gamma_0/n_{2\omega}]\cos 2\varphi + [M_0n_1n_{2\omega}b_1b_5 + n_1b_3\gamma_0/n_{2\omega}]\sin 2\varphi\}]\cdot[n_1^2(b_3^2+b_5^2) + (1-M_0^2)(n_{2\omega}^2b_1^2/2 + \gamma_0^2/(2n_{2\omega}^2) - n_1n_{2\omega}b_1b_3) + \sqrt{1-M_0^2}\,\{[n_1^2(b_3^2+b_5^2) - n_1n_{2\omega}b_1b_3 + n_1b_5M_0\gamma_0/n_{2\omega}]\cos 2\varphi + [n_1n_{2\omega}b_1b_5 + n_1b_3M_0\gamma_0/n_{2\omega}]\sin 2\varphi\}]^{-1} \tag{6}$$

$$\Psi(\varphi) = 0.5\mathrm{Arg}\left\{ \left([n_1(b_3 - \mathrm{i}b_5) - n_{2\omega}b_1]^2 + \gamma_0^2/n_{2\omega}^2\right)\cdot\exp(2\mathrm{i}\varphi) + n_1^2(b_3 - \mathrm{i}b_5)^2\cdot\exp(-2\mathrm{i}\varphi) + \frac{2n_1(b_3-\mathrm{i}b_5)}{\sqrt{1-M_0^2}}[n_1(b_3-\mathrm{i}b_5) - n_{2\omega}b_1 + \mathrm{i}M_0\gamma_0/n_{2\omega}] \right\} \tag{7}$$

In formulas (5), (6)

$$D(r,z) = \frac{1}{\beta^2(z)}\frac{16\sqrt{2}\pi \mathrm{i}E_0^2\exp(-2\mathrm{i}\omega t - \mathrm{i}k_2 z)}{\omega w^2(1+n_1)^2(1+n_{\mathrm{SH}})} r \times \exp\{-2r^2/w^2\beta(z)\},$$

$x = r\cos\varphi$, $y = r\sin\varphi$. We should notice, that formula (7) makes any sense only if $M_0 \neq \pm 1$.

It can be seen from (5)–(7), that M and ψ do not depend on the propagation coordinate z and the transversal coordinate r. The expression for the angle Φ between the electric field vector and the x-axis of the coordinate system at fixed timing for arbitrary values of z is very cumbersome and non-informative. This fact is circumstanced by the parabolic phase accumulation due to the traveling in the linear medium. At $z = 0$ this expression has the following form:

$$\Phi(\varphi, z=0) = \mathrm{Arg}\{\sqrt{1-M_0^2}\cdot(\gamma_0/n_{2\omega})\cdot\exp(\mathrm{i}\varphi) + \mathrm{i}M_0 n_1(b_3 - \mathrm{i}b_5)\exp(-\mathrm{i}\varphi)\} \tag{8}$$

3. Discussion of results

The expressions (5)–(8) give full information about the distributions of intensity and polarization in the transversal section of the SH beam. It can be seen from these, that $I(\varphi)$, $M(\varphi)$, $\psi(\varphi)$ $\Phi(\varphi)$ are, of course, the periodic functions, but $I(\varphi)$, $M(\varphi)$, and $\psi(\varphi)$ have a period of π, and $\Phi(\varphi)$ has a period of 2π. It is easy to show that the directions of the electric field vector in any two diametrically opposite points of the cross-section of the beam at fixed timing are opposite to each other. That is why the polarization structure of the SH beam is symmetric to 180°-rotation. Taking this in mind, we analyze $I(\varphi)$, $M(\varphi)$, $\psi(\varphi)$ and $\Phi(\varphi)$ only for $0 \leqslant \varphi \leqslant \pi$.

All the four quantities (5)–(8) essentially depend on the constants characterizing the spatial dispersion of quadratic optical response of the nonlinear medium and the surface nonlinearity, and also on the polarization of the incident radiation.

Fig. 1a shows the example of inhomogeneous distribution of the intensity and the polarization of the SH light field (in dimensionless units $x_1 = 2x/w$ and $y_1 = 2y/w$) in terms of the polarization ellipses in separate points of the cross-section of the SH beam at $z = 0$ (nearby the surface) in case of elliptic polarization of the incident radiation. The sum of squared axes of each ellipse is proportional to the intensity of light in the point, given by the center of the ellipse (at radius-vector r), the relation of the axes of the ellipse can be unambiguously expressed through the $M(\mathbf{r})$, and the inclination angle of the main axis of the ellipse is equal to $\psi(\mathbf{r})$. The orientation of the electric field vector at fixed timing in the point given by $\mathbf{r}$ is shown by the small circle at the boundary of the ellipse. Shaded ellipses correspond to the clockwise rotation of the polarization vector and opened ellipses correspond to the counterclockwise rotation. It can be seen from the figure that the polarization state essentially changes along the beam cross-section, and one can find regions with linear ($M(r,\varphi) = 0$), elliptical ($-1 < M(r,\varphi) < 1$) and circular ($M(r,\varphi) = \pm 1$) polarizations. For comparison, the transversal polarization distribution of the fundamental beam is shown in Fig. 1b. Unlike in Fig. 1a, all the ellipses are oriented in the same way and have the same ellipticity degree, and $\Phi(\mathbf{r}, z) \equiv 0$.

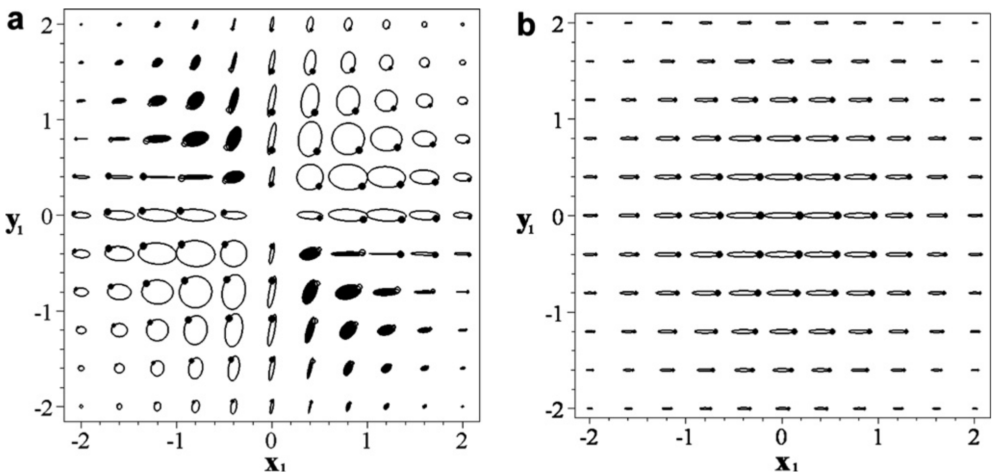

Fig. 1. The transversal spatial distributions of light polarization in the SH beam (a) and in the fundamental beam (b). $M_0 = 0.3$, $n_1 = 1.33$, $n_{2\omega} = 1.35$, $b_3/b_1 = 1.2$, $b_5/b_1 = 0.2$, $\gamma_0/b_1 = 2$.

Using (5)–(8) it is not difficult to find that in the case of the absence of a nonlocal bulk response of the medium ($\gamma_0 = 0$), dependencies $I(M_0)$, $\psi(M_0)$ (for arbitrary fixed value of φ) become even functions, and $M(M_0)$ becomes an odd function. The value of $\Phi(M_0)$ suffers a shift on π if the sign of M_0 is changed. It is worth to notice that if the fundamental wave is linearly polarized in this case (i.e. $M_0 = 0$, $\gamma_0 = 0$), then the SH radiation is also linearly polarized in each point of the SH beam cross-section. At that, the phase difference between the oscillations of the electric field vector in each of the two points of the cross-section can be equal either to zero or to π. Also if $\gamma_0 = 0$, the $I(\varphi)$ and $M(\varphi)$ dependencies have the same coordinates $\varphi_{\max}$ and $\varphi_{\min}$ for minimum and maximum.

If there exists a nonlocal response of the medium bulk, the dependence of the polarization state of the SH radiation becomes more complex. In this case the functions $I(M_0)$, $\psi(M_0)$ and $M(M_0)$ do not possess even or odd properties, but the simultaneous change of the sign of M_0 и γ_0 preserves the values of $I(M_0, \gamma_0)$ and $\psi(M_0, \gamma_0)$ and their sign, and changes the sign of $M(M_0, \gamma_0)$ (preserving its absolute value). The value of $\Phi(M_0, \gamma_0)$ suffers a shift on π under this condition. The coordinates of maxima and minima of $I(\varphi)$ and $M(\varphi)$ are different. Even if the fundamental wave is linearly polarized, the SH radiation will have an elliptical polarization in general case, and the polarization state will be inhomogeneously distributed along the SH beam cross-section.

Fig. 2 demonstrates the behavior of the spatial distribution of the polarization of light at double frequency near the surface of the medium for different values of the ellipticity degree of the incident wave polarization ellipse and for fixed values of medium parameters: $n_1 = 1.33$, $n_{2\omega} = 1.35$, $b_3/b_1 = 1.2$, $b_5/b_1 = 0.2$, $\gamma_0/\mathrm{b}_1 = 2$. In Fig. 2a each ellipse in the horizontal line corresponding to the given M_0 (marked at ordinate-axis) shows the polarization state and the electric field vector orientation at fixed timing at the radial direction corresponding to the polar angle φ (marked at abscissa axis) in the beam cross-section. Fig. 2b–d shows $\psi(\varphi)$, $\Phi(\varphi)$ and $M(\varphi)$ for different values of M_0. The range of variation of the angle of rotation $\psi(\varphi)$ (Fig. 2b) achieves maximum size, when M_0 is close to zero. $M(\varphi)$ changes very slightly if $|M_0|$ is close to 1 (Fig. 2d). Apart from this, it can be seen from Fig 2a and d that both intensity extrema as well as both ellipticity degree extrema are located at mutually perpendicular radial directions in the beam cross-section (this also directly follows from (5) and (6)).

When the incident beam is circularly polarized, the reflected beam at double frequency is also circularly polarized with the same direction of polarization rotation as in the incident beam. In this case the transversal distribution of intensity of the SH radiation has radial symmetry and is proportional to $(|b_3|^2 + |b_5|^2)/|b_1|^2$, i.e. the signal wave in this case depends only on the surface nonlinearity.

In general case, when $M_0 \neq 0$, the equation $M(\varphi) = 0$ may have two roots, one root or no real roots. In the first case the cross-section of the SH beam is divided by two direct lines $\varphi = \varphi_{1,2}$ (roots of the equation $M(\varphi) = 0$) crossing its center into four sectors in such a way that the directions of rotation of the polarization vector in each of the two neighboring sectors are opposite, because the ellipticity degree changes its sign at these lines (such case is illustrated at Fig. 1a). These lines are given by the following expressions:

$$\varphi_{1,2} = \mathrm{arctg}[(-B \pm \sqrt{B^2 - AC})/A)], \tag{9}$$

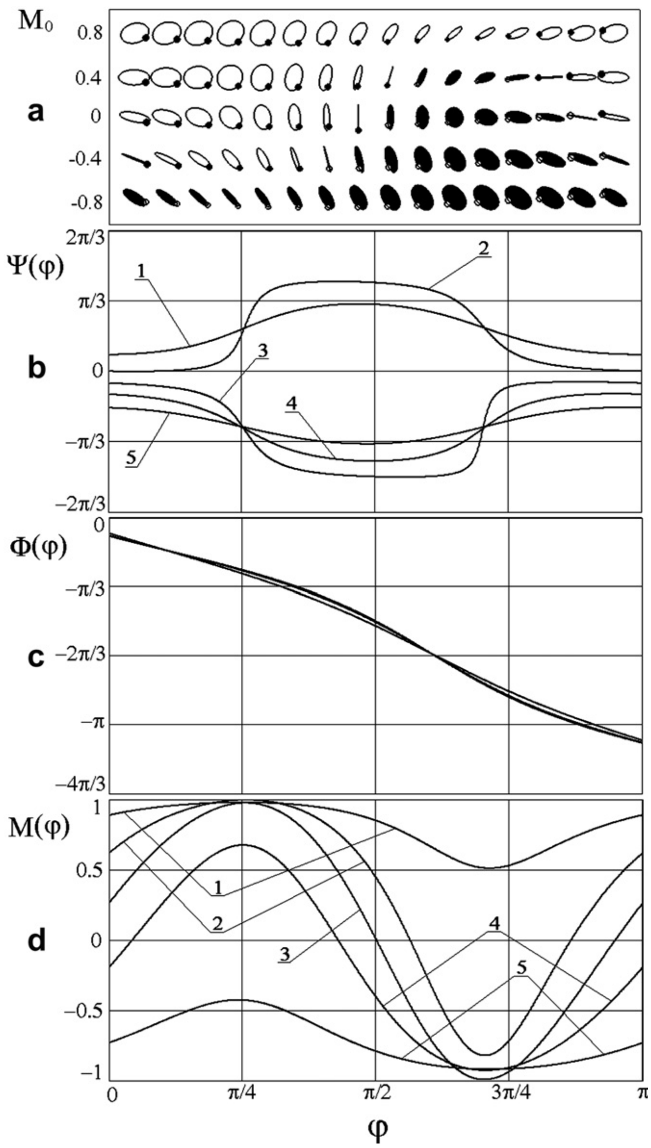

Fig. 2. The dependencies of (a) the shape and orientation of the polarization ellipse, (b) the angle of polarization ellipse rotation, (c) the orientation of the electric field vector at fixed timing and (d) the ellipticity degree of the polarization ellipse on the polar angle coordinate in the cross-section of the SH beam. Curves 1–5 correspond to $M_0 = -0.8$, -0.4, 0, 0.4, 0.8 (curves at (c) almost coincide with each other). $n_1 = 1.33$, $n_{2\omega} = 1.35$, $b_3/b_1 = 1.2$, $b_5/b_1 = 0.2$, $\gamma_0/b_1 = 2$.

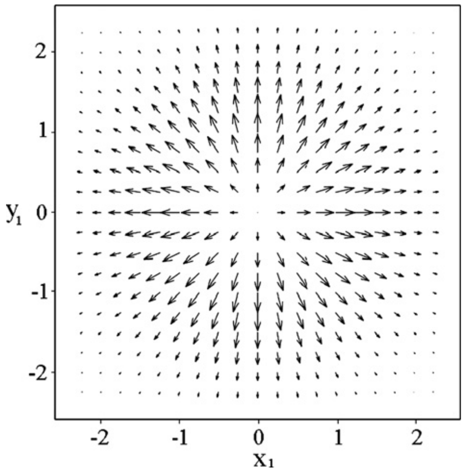

Fig. 3. Radially polarized SH beam, generated in the medium with $b_3 = 0$, $b_5 = 0$ irrespective of the fundamental wave polarization. $n_{2\omega} = 1.35$, $n_1 = 1.33$; $\gamma_0/b_1 = 1$.

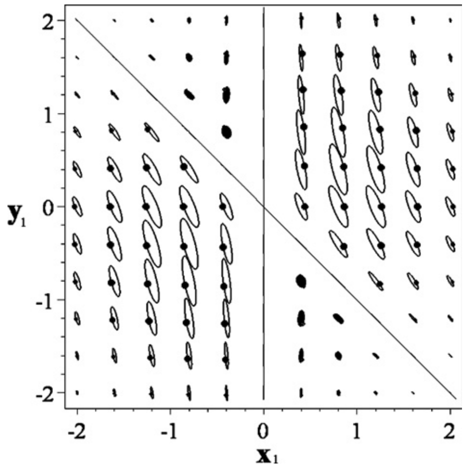

Fig. 4. Polarization distribution in the SH beam cross-section in the case of linear polarization of fundamental wave $M_0 = 0$ and the following values of medium parameters: $b_3/b_1 = b_5/b_1 = 2$; $n_1 = 1.33$; $n_{2\omega} = 1.35$; $\gamma_0/b_1 = 0.5$. Lines marked at the figure correspond to roots of the equation $M(\varphi) = 0$. The SH radiation is linearly polarized at these lines, and the polarization plane is oriented along them.

where $A = (1 - \sqrt{1 - M_0^2})(M_0 n_1 (b_3^2 + b_5^2) + b_5 \gamma_0 / n_{2\omega}) - M_0 (M_0 b_5 \gamma_0 / n_{2\omega} + \sqrt{1 - M_0^2} \cdot n_{2\omega} b_1 b_3)$, $B = \sqrt{1 - M_0^2} (b_3 \gamma_0 / n_{2\omega} + n_{2\omega} b_1 b_5 M_0)$, $C = M_0 n_1 (b_3^2 + b_5^2) + (1 - M_0^2) b_5 \gamma_0 / n_{2\omega}$.

If $B^2 - AC = 0$ then the SH radiation is linearly polarized at the direct line $\varphi = \varphi_0$, where $\varphi_0 = \mathrm{arctg}[-B/A]$, and the direction of rotation of the polarization vector does not change in the beam cross-section. And, finally, in the third case, when $B^2 - AC < 0$, there is only an elliptically polarized radiation at double frequency and the direction of the polarization rotation does not change in the SH beam.

The following two particular cases of the considered problem are of special interest because these are connected with possible spectroscopic applications.

If the nonlinear optical response of the surface can be neglected $\hat{\kappa}^{(2)} \equiv 0$, then for any values of M_0 the SH radiation is linearly polarized in each point of the reflected beam and the polarization plane is oriented along the radius of the beam in each point: $\Psi(\varphi)|_{\hat{\kappa}^{(2)}=0} = \varphi$. Such a radial polarization distribution in the SH beam is shown at Fig. 3. The length of each arrow at the figure is proportional to the amplitude of the light field in the corresponding point of the beam cross-section. The orientation of the arrow (equal to the orientation of the polarization plane) coincides with the polar angle φ in each point.

It can be shown from (9), that if the fundamental beam is linearly polarized, then there are the lines $\varphi_1 = \pi/2$ and $\varphi_2 = -\mathrm{arctg}(b_5/b_3)$ in the SH beam cross-section, where the radiation is linearly polarized (these lines $\varphi_{1,2}$ correspond to the roots of the equation $M(\varphi) = 0$), and the plane of polarization at these lines is oriented along them, i.e. $\psi(\varphi = \pi/2;\ M_0 = 0) = \pi/2$, $\psi(\varphi = -\mathrm{arctg}(b_5/b_3);\ M_0 = 0) = -\mathrm{arctg}(b_5/b_3)$. Fig. 4 illustrates such a case and the corresponding lines are marked there; $b_5/b_3 = 1$ and the inclination angle of the second line is equal to $-45°$. Therefore, detecting the part of the SH beam, which is close to the certain line crossing the center of the beam (for example, by means of slit aperture), and choosing this line in such a way that the polarization of the detected light is linear and the polarization plane is oriented along the selected line, we measure the positions of these lines and the angle between them. The result of our measurement will give us the value of b_5/b_3. Further measurement of the intensity of the SH radiation in the case of circularly polarized pump wave will give us a combination $(|b_3|^2 + |b_5|^2)/|b_1|^2$, which together with b_5/b_3 allows to simply find b_5/b_1 and b_3/b_1.

For experimental observation of the polarization effects described above in the cross-section of the reflected beam at double frequency (for example, the search of directions in the beam, where light is linearly polarized, and the polarization plane is oriented along the radius) it is sufficient to use the method described in [35], where the polarization effects in the beam cross-section were investigated for the problem of light self-action: the radiation reflected from the nonlinear medium passes through the polarizer, and the resulting distribution of light intensity is measured on the CCD-matrix. By rotating the polarizer with sufficiently small angle step and by measuring the intensity distribution at CCD-matrix for all the positions of the polarizer, we can find the polarization of light in every point of the beam cross-section, recording the values of maximum and minimum intensity detected in the given point for all the orientations of the polarizer and the angle of polarizer orientation corresponding to maximum. If necessary, the beam falling on the CCD-matrix can be resized by means of lens or system of lenses. If one needs to measure the intensity distributions of the circularly polarized light components with either right or left rotation directly, the birefringent plate "$\lambda/4$" can be installed before the polarizer, as in [35]. In this case the intensity distribution of the corresponding component of light can be obtained by orienting the polarizer at either +45° or −45° for detecting the right circularly polarized or the left circularly polarized component. This provides an easy calculation of the ellipticity degree distribution.

4. Conclusion

In this work we present the detailed analytical study of the polarization effects in the cross-section of the SH beam generated from the surface of a chiral medium by a normally incident focused Gaussian beam of a fundamental radiation. The polarization properties of SH beam depend only on the polar angle coordinate in the cross-section of the beam, and the coordinate dependencies of the intensities of the circularly polarized light components are the products of polar radius dependence and polar angle dependence. This fact essentially simplifies the analysis of these distributions and allows one to acquire the information about the medium nonlinearity by selective detection of the radiation from the certain areas of the SH beam cross-section (along certain lines crossing the center of the beam) and by subsequent measurement of the polarization state of light in these areas and also by search of the radial directions in the beam cross-section, which correspond to maximum and minimum of intensity.

It is shown that in the case of the absence of surface contribution to the signal at double frequency or its negligibility compared to bulk contribution, the SH beam becomes radially polarized for any polarization state of the incident beam, and the intensity of SH is maximum if the incident beam is linearly polarized.

The authors believe that the results obtained in this work are promising for future applications for spectroscopy analysis of surfaces of chiral medium or thin chiral layers and for the methods of formation of inhomogeneously polarized light beams. The authors are grateful to Dr. S.N. Volkov for valuable discussions.

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