

LASER BEAMS AND RESONATORS

PACS numbers: 42.65.Ky; 42.25.Ja
DOI: 10.1070/QE2006v036n09ABEH013269

Formation of an inhomogeneously polarised light beam at the sum frequency by two collinear elliptically polarised Gaussian beams focused into a chiral medium

S.N. Volkov, V.A. Makarov, I.A. Perezhogin

***Abstract*. The distribution of polarisation of a light field in the cross section of a beam at the sum frequency is investigated upon the collinear interaction of two elliptically polarised Gaussian beams in a nonlinear isotropic gyrotropic medium. It is shown that the ellipticity, the angle of rotation of the principal axis of the polarisation ellipse, and the rotation direction of the electric field vector of radiation at the sum frequency in the beam cross section strongly depend on the angle in the polar coordinate system. The ranges of parameters of elliptically polarised fundamental Gaussian beams are found where the cross section of the sum-frequency beam is divided into sectors with different rotation directions of the electric field vector. The equations of the straight lines determining the boundaries of these sectors contain parameters specifying the shape and orientation of polarisation ellipses of the fundamental waves and the ratio of their wave vectors. In the case of opposite circular polarisations of these waves, the ellipticity of the sum-frequency beam does not change in the beam cross section and the principal axes of polarisation ellipses of the light field are oriented perpendicular to the radius in polar coordinates.**

Keywords: *sum-frequency generation, elliptic polarisation, Gaussian beam, spatial dispersion, gyrotropy.*

1. Introduction

The polarisation self-action and interaction of light beams is described, as a rule, by obtaining the system of nonlinear equations for slowly varying complex amplitudes of linearly or circularly polarised orthogonal components of the light field [1]. The solution of this system allows one to analyse [2] variations in the intensity $I(\boldsymbol{r}, z)$, the ellipticity $M(\boldsymbol{r}, z)$, the angle of rotation $\Psi(\boldsymbol{r}, z)$ of the principal axis of the polarisation ellipse, and the angle $\alpha(\boldsymbol{r}, z)$ determining the orientation of the electric field vector at a fixed instant of time (measured, for example, from the principal axis of the polarisation ellipse) at different points of the cross section of a light beam propagating along the z axis ($\boldsymbol{r}$ is the radius-vector component in the xy plane).

Different distributions of the light-field polarisation in the plane perpendicular to the beam propagation axis can be most conveniently and clearly illustrated by polarisation ellipses constructed at different points of the beam cross section [2]. The sum of squares of the semiaxes of the polarisation ellipse is proportional to the light intensity at its centre, the axial ratio is uniquely determined by the parameter $M(\boldsymbol{r}, z)$, and the tilt angle of its principal axis is equal to the angle $\Psi(\boldsymbol{r}, z)$. The orientation $\alpha(\boldsymbol{r}, z)$ of the electric field vector at the fixed instant of time characterises the phase of its oscillations.

Theoretical and experimental studies performed to date have shown conclusively that the polarisation self-action and interaction of waves are delicate but widespread effects of nonlinear optics [3]. The polarisation of waves incident on a nonlinear medium substantially determines processes of nonlinear optical interaction and self-action of light. Therefore, a time-consuming consideration of a change in the polarisation of the interacting waves is justified and is of interest. However, this consideration is performed, as a rule, in the plane wave approximation. In the case of beams, only linearly polarised waves incident on a nonlinear medium are usually considered or a change in the elliptical polarisation near their axes with increasing propagation coordinate is analysed [4, 5]. This is explained not only by cumbersome expressions obtained for $I(\boldsymbol{r}, z)$, $M(\boldsymbol{r}, z)$, $\Psi(\boldsymbol{r}, z)$, and $\alpha(\boldsymbol{r}, z)$ in the problems of nonlinear optics (which are sometimes represented in quadratures) and difficulties involved in the interpretation of the found dependences but also by the absence of stimulating experiments. In addition, many nonlinear effects of the frequency shift are forbidden in the plane wave approximation [6] but are quite possible if the spatial limitedness of light beams is taken into account [4], and therefore the possibility of the appearance of a signal wave is discussed first of all.

The above discussion concerns to a great extent the problem of sum-frequency generation in an isotropic chiral medium (the symmetry $\infty\infty$) by two focused coaxial copropagating Gaussian beams sharing the waist plane [7]. The appearance of a sum-frequency signal in this case is related to the local electric dipole optical susceptibility $\hat{\chi}^{(2)}(\omega_1 + \omega_2; \omega_1; \omega_2)$ and is forbidden in the plane wave approximation because the polarisation vector of the medium at the sum frequency produced by two plane pump waves with parallel wave vectors has only the longitudinal

S.N. Volkov Department of Physics, University of Toronto, 60 St. George Street, Toronto, Ontario, M5S 1A7, Canada;
V.A. Makarov Department of Physics and International Laser Center, M.V. Lomonosov Moscow State University, Vorob'evy gory, 119992 Moscow, Russia; e-mail: vamakarov@phys.msu.ru;
I.A. Perezhogin Department of Physics, M.V. Lomonosov Moscow State University, Vorob'evy gory, 119992 Moscow, Russia;
e-mail: iap1@mail.ru

Received 13 March 2006; revision received 6 June 2006
Kvantovaya Elektronika **36** (9) 860 – 866 (2006)
Translated by M.N. Sapozhnikov

component and cannot be the source of a free transverse signal wave. Spatially limited beams have small (of the first order of smallness in the divergence angle) longitudinal components of the electromagnetic field [to satisfy the condition $\operatorname{div}\boldsymbol{E}(\omega_{1,2}) = 0$ in vacuum], which make possible the experimental observation of sum-frequency generation in this geometry.

Sum-frequency generation in a chiral medium by two focused coaxial Gaussian beams was considered in [7]. This problem is of current interest for revealing mechanisms that can be responsible for second-harmonic generation in an isotropic non-centrally symmetric suspension of fragments of purple membranes of bacteria *Halobacterium halobium* observed during the propagation of a linearly polarised laser pulse in the suspension [6].

In [7], quadrature formulas for the electric field strength and the wave power at the sum frequency were obtained and analytic expressions were found for these quantities in the case of the exact phase synchronism and tight focusing of fundamental beams to the centre of an extended medium. It was shown that in the latter case the sum-frequency generation is impossible if $\Delta k = k_1 + k_2 - k_{\rm sf} < 0$ (where k_1, k_2, and $k_{\rm sf}$ are the wave numbers of elliptically polarised electromagnetic waves with frequencies ω_1, ω_2, and $\omega_{\rm sf} = \omega_1 + \omega_2$, respectively, propagating in a medium), while for some positive Δk quasi-phase matching is achieved. In the latter case, the transverse intensity distribution of the beam at the sum frequency has the form of a ring with radius increasing with Δk. The dependence of the wave power at the sum frequency on the polarisation of fundamental beams was also thoroughly studied. The results obtained in [7] have demonstrated that such interaction geometry of fundamental beams can be promising for obtaining quasi-phase matching.

The spatial distribution of polarisation of the light field in a plane perpendicular to the propagation direction of the signal wave at the sum frequency was studied neither in classical paper [8] nor later (see, for example, [9–14]). The only exclusion is the field distribution in the beam cross section at the sum frequency obtained in [7] for the simplest case of linearly polarised fundamental waves.

In this paper, we studied the formation of a light beam inhomogeneously polarised in the cross section in an isotropic chiral medium upon generation of the sum frequency by two focused collinear homogeneously elliptic polarised Gaussian beams. The images of polarisation ellipses at different points of the light beam cross section at the sum frequency constructed by using the formulas obtained illustrate strong dependences of the intensity, parameters of the polarisation ellipse of the light field, and the orientation angle $\alpha(\boldsymbol{r}, z)$ of the electric field vector at a fixed instant on transverse coordinates. A complicated type of the dependence $\alpha(\boldsymbol{r}, z = \text{const})$ is caused by a strongly varying phase difference of electric-field oscillations at different points of the light beam cross section at a fixed instant of time. Note that in the plane wave approximation, $\alpha(\boldsymbol{r}, z) \equiv 0$ for any $\boldsymbol{r}$ and z.

2. Formulation of the problem and its analytic solution

The concepts of the 'transverse' and 'longitudinal' components of the electric field of an electromagnetic wave are convenient only in the case of plane waves. The beam can be naturally represented as a superposition of the plane-wave spatial Fourier harmonics whose wave vectors are slightly noncollinear, and therefore even in the case when the field in the beam is polarised perpendicular to its axis, it should have nevertheless a small longitudinal component. The natural generalisation of these two concepts is the beams of the so-called vortex and potential types. They are specified by the conditions $\operatorname{div}\boldsymbol{E} = 0$ and $\operatorname{rot}\boldsymbol{E} = 0$, respectively. One can easily see that in this case of such a definition, each spatial Fourier harmonic of the potential or vortex beam is the longitudinal or transverse plane wave, respectively, and, therefore, all the specific properties of longitudinal or transverse electromagnetic waves will be 'inherited' by the potential or vortex beams, respectively.

Note that the potential beam has a small field component directed perpendicular to its axis, while the vortex beam has a small longitudinal component. These components are the quantities of the first order of smallness in the divergence angle of the beam. It is known that the potential beam cannot propagate freely and exits only inside a medium, being 'coupled' with the corresponding polarisation wave of matter. Having approached the surface, it makes, due to boundary conditions, the contribution to a free wave; however, this contribution should be taken into account as a part of the signal from the surface. In this paper, as in [5, 7], we will consider only vortex electromagnetic waves at the sum frequency because only these waves are produced in the medium.

Let us assume that the symmetry axes of both homogeneously elliptically polarised fundamental Gaussian beams with $\boldsymbol{E}_m(\boldsymbol{r})$ ($m = 1, 2$) propagating in a nonlinear isotropic gyrotropic medium coincide with the z axis and the beams share the waist plane at $z = l_0$. Then,

$$\boldsymbol{E}_m(\boldsymbol{r}) = [\boldsymbol{e}_m + \mathrm{i}k_m^{-1}\boldsymbol{e}_z(\boldsymbol{e}_m\nabla)] \times \frac{E_{0m}}{\beta_m(z)} \exp\left[-\mathrm{i}\omega_m t + \mathrm{i}k_m(z - l_0) - \frac{r^2}{w_m^2\beta_m(z)}\right]. \quad (1)$$

Here, $\nabla = \partial/\partial\boldsymbol{r}$; $\beta_m(z) = 1 + \mathrm{i}(z - l_0)/l_{{\rm d}m}$; $l_{{\rm d}m} = k_m w_m^2/2$ is the diffraction length; w_m is the beam waist half-width; E_{0m} is the scalar complex amplitude; $\boldsymbol{e}_m$ is the complex polarisation vector of the beam with frequency ω_m ($|\boldsymbol{e}_m(\boldsymbol{r}, t)|^2 = 1$); and $\boldsymbol{e}_z$ is the unit vector along the z axis. Note that expression (1) satisfies the equation $\operatorname{div}\boldsymbol{E} = 0$ with an accuracy to the term of the first order of smallness in the divergence angle. We assume that the wave number k_m is the same for the right- and left-hand circularly polarised waves, thereby neglecting linear absorption and linear gyration. The consideration of the latter gives rise only to small corrections to rather complex analytic formulas.

The solution of the parabolic equation for the slowly varying amplitude $\boldsymbol{A}$ of the vortex component $\boldsymbol{E}_\perp^{\rm sf} = \boldsymbol{A}\exp[-\mathrm{i}\omega_{\rm sf}t + \mathrm{i}k_{\rm sf}(z - l_0)]$ of the field of the signal wave at the sum frequency, which contains in the right-hand side the vortex component of the polarisation vector $\boldsymbol{P}(\omega_{\rm sf}; \boldsymbol{r}) = \chi^{(2)}[\boldsymbol{E}_1(\boldsymbol{r})\boldsymbol{E}_2(\boldsymbol{r})]$ of matter, was found in quadratures in [7]:

$$\boldsymbol{A}(\boldsymbol{r}, z) = \sqrt{2}F_0(r, z)\left\{(\boldsymbol{e}_1\boldsymbol{r})[\boldsymbol{e}_z\boldsymbol{e}_2] + \frac{k_1}{k_{\rm sf}}\,\boldsymbol{r}(\boldsymbol{e}_z[\boldsymbol{e}_1\boldsymbol{e}_2])\right\}, \quad (2)$$

where

$$F_0(r,z) = \frac{\sqrt{2}\pi i k_{sf}\chi^{(2)}E_{01}E_{02}(l_{d2}-l_{d1})l_s}{\varepsilon_{sf}l_{d1}l_{d2}[1+i(z-l_0)/l_s]^2}\,J(r,-l_0,z-l_0); \quad (3)$$

ε_{sf} is the dielectric constant of matter at the frequency $\omega_1+\omega_2$; $\chi^{(2)} = \chi^{(2)}_{xyz}(\omega_{sf};\omega_1;\omega_2)$ is the only independent component of the tensor of quadratic local optical susceptibility of the medium; and $l_s = (k_1+k_2)w_s^2/2$; $w_s^{-2} = w_1^{-2}+w_2^{-2}$. The dimensionless integral $J(z)$ determines the dependence of $\boldsymbol{E}_\perp^{sf}$ on the propagation coordinate

$$J(r,-l_0,z-l_0\equiv l_s\zeta) = \int_{-l_0/l_s}^{\zeta} d\zeta'\,\frac{(1+i\zeta)^2}{B^2(\zeta',\zeta)}$$

$$\times\exp\left[i v_{sf}\zeta' - \frac{r^2}{w_s^2}\,\frac{1+il_s^2\zeta'/(l_{d1}l_{d2})}{B(\zeta',\zeta)}\right], \quad (4)$$

where $\zeta = (z-l_0)/l_s$ is the normalised propagation coordinate; $v_{sf} = \Delta k l_s$; Δk is the mismatch of the wave vectors determined earlier; and

$$B(\zeta',\zeta) = \left(1+i\frac{l_s}{l_{d1}}\zeta'\right)\left(1+i\frac{l_s}{l_{d2}}\zeta'\right)$$

$$-i\,\frac{2l_s}{k_{sf}w_s^2}(\zeta'-\zeta)\left(1+i\frac{l_s\zeta'}{l_{d1}l_{d2}}\right). \quad (5)$$

The field $\boldsymbol{E}^{sf}$ is completely characterised by the normalised intensity $I(\boldsymbol{r},z) = |\boldsymbol{A}|^2$, the ellipticity $M(\boldsymbol{r},z) = 2\,\mathrm{Im}(A_xA_y^*)/|\boldsymbol{A}|^2$ (note that $-1\leqslant M\leqslant 1$), the angle of rotation of the principal axis of the polarisation ellipse

$$\Psi(\boldsymbol{r},z) = 0.5\arg\{|A_x|^2-|A_y|^2+2i\mathrm{Re}(A_xA_y^*)\}, \quad (6)$$

and the angle

$$\alpha(\boldsymbol{r},z,t) = \arg\{\cos[\theta(\boldsymbol{r},z,t)] + i\gamma(\boldsymbol{r},z)\sin[\theta(\boldsymbol{r},z,t)]\}, \quad (7)$$

which determined the orientation of the electric filed vector at the instant t measured, for example, from the principal axis of the polarisation ellipse. In expression (7),

$$\theta(\boldsymbol{r},z,t) = 0.5\arg\{(A_x^2+A_y^2)$$

$$\times\exp[-2i\omega_{sf}t+2ik_{sf}(z-l_0)]\}, \quad (8)$$

$$\gamma(\boldsymbol{r},z) = \frac{[1+M(\boldsymbol{r},z)]^{1/2}-[1-M(\boldsymbol{r},z)]^{1/2}}{[1+M(\boldsymbol{r},z)]^{1/2}+[1-M(\boldsymbol{r},z)]^{1/2}}. \quad (9)$$

The dependence of $\tilde\alpha(\boldsymbol{r},z) = \alpha(\boldsymbol{r},z,t=k_{sf}(z-l_0)/\omega_{sf})$ on $\boldsymbol{r}$ describes a change in the field oscillation phase at different points of the beam cross section, while the parameter $|\gamma|$ is equal to the axial ratio of the polarisation ellipse at the point with coordinates $\boldsymbol{r}$ and z. The end of the electric field vector moves not uniformly over the polarisation ellipse with the angular velocity

$$\dot\alpha(\boldsymbol{r},z,t) = \frac{-\gamma(\boldsymbol{r},z)\omega_{sf}}{\cos^2[\theta(\boldsymbol{r},z,t)]+\gamma(\boldsymbol{r},z)^2\sin^2[\theta(\boldsymbol{r},z,t)]}. \quad (10)$$

In this case, the value of $\dot\alpha(\boldsymbol{r},z,t)$ averaged over period is ω. Note that the rotation direction of the electric field vector [the sign of $\dot\alpha(\boldsymbol{r},z,t)$] is determined by the sign of $M(\boldsymbol{r},z)$. By using (2)–(5), we can easily obtain that

$$I(r,\varphi,z) = |rF_0(\boldsymbol{r},z)|^2\big(1-\kappa(1-\kappa)$$

$$\times\big\{1-\big[(1-M_{01}^2)(1-M_{02}^2)\big]^{1/2}\cos 2\Psi_2 - M_{01}M_{02}\big\}$$

$$+\mathrm{Re}\big\{\exp(2i\varphi)\big[\kappa\exp(-2i\Psi_2)(1-M_{02}^2)^{1/2}$$

$$+(1-\kappa)(1-M_{01}^2)^{1/2}\big]\big\}\big), \quad (11)$$

$$M(\varphi) = -\big(M_{01}+(1-\kappa)M_{02}+\mathrm{Re}\big\{\exp(2i\varphi)$$

$$\times\big[\kappa\exp(-2i\Psi_2)M_{01}(1-M_{02}^2)^{1/2}$$

$$+(1-\kappa)M_{02}(1-M_{01}^2)^{1/2}\big]\big\}\big)\big(1-\kappa(1-\kappa)\big\{1-M_{01}M_{02}$$

$$-\big[(1-M_{01}^2)(1-M_{02}^2)\big]^{1/2}\cos 2\Psi_2\big\}+\mathrm{Re}\big\{\exp(2i\varphi)$$

$$\times\big[\kappa\exp(-2i\Psi_2)(1-M_{02}^2)^{1/2}+(1-\kappa)(1-M_{01}^2)^{1/2}\big]\big\}\big)^{-1}, \quad (12)$$

$$\Psi(\varphi) = 0.5\arg\big\{-2\big[(1-M_{02}^2)^{1/2}\exp(2i\Psi_2)(1-\kappa)$$

$$+(1-M_{01}^2)^{1/2}\kappa\big]+\exp(2i\varphi)\big\{\kappa^2\big[(1+M_{01})(1-M_{02})$$

$$-2\big[(1-M_{01}^2)(1-M_{02}^2)\big]^{1/2}\cos 2\Psi_2+(1-M_{01})(1+M_{02})\big]$$

$$-\kappa\big[(1+M_{01})(1-M_{02})-2\big[(1-M_{01}^2)(1-M_{02}^2)\big]^{1/2}$$

$$\times\exp(2i\Psi_2)+(1-M_{01})(1+M_{02})\big]$$

$$-\big[(1-M_{01}^2)(1-M_{02}^2)\big]^{1/2}\exp(2i\Psi_2)\big\}-\exp(-2i\varphi)$$

$$\times\big[\big[(1-M_{01}^2)(1-M_{02}^2)\big]^{1/2}\exp(2i\Psi_2)\big]\big\}, \quad (13)$$

$$\tilde\alpha(r,\varphi,z) = \arg\big\{0.5\exp(-i\varphi)\big\{\big[(1+M_{01})(1+M_{02})\big]^{1/2}$$

$$\times\exp(2i\Psi_2)rF_0(r,z)-\big[(1-M_{01})(1-M_{02})\big]^{1/2}rF_0^*(r,z)\big\}$$

$$+i\exp(i\varphi)\big\{\kappa\big(\big[(1+M_{01})(1-M_{02})\big]^{1/2}\mathrm{Im}[rF_0(r,z)]$$

$$-[(1-M_{01})(1+M_{02})]^{1/2}\mathrm{Im}[rF_0(r,z)\exp(2i\Psi_2)]\big)$$

$$+[(1-M_{01})(1+M_{02})]^{1/2}\mathrm{Im}[rF_0(r,z)\exp(i\Psi_2)]\exp(i\Psi_2)\big\}\big\}$$

$$-0.5\arg\big\{-2\big[(1-M_{02}^2)^{1/2}\exp(2i\Psi_2)(1-\kappa)$$

$$+(1-M_{01}^2)^{1/2}\kappa\big]+\exp(2i\varphi)$$

$$\times\big\{2\kappa^2\big[1-M_{01}M_{02}-[(1-M_{01}^2)(1-M_{02}^2)]^{1/2}\cos 2\Psi_2\big]$$

$$-2\kappa\big[1-M_{01}M_{02}-[(1-M_{01}^2)(1-M_{02}^2)]^{1/2}\exp(2i\Psi_2)\big]$$

$$-[(1-M_{01}^2)(1-M_{02}^2)]^{1/2}\exp(2i\Psi_2)\big\}$$

$$-\exp(-2i\varphi)\big\{[(1-M_{01}^2)(1-M_{02}^2)]^{1/2}\exp(2i\Psi_2)\big\}\big\}, \quad (14)$$

where φ is the polar angle determining the direction of the vector $\boldsymbol{r}$ ($0 < \varphi < 2\pi$); Ψ_2 is the angle between the principal axes of polarisation ellipses of the fundamental waves at the input to the medium, which is the same at all the points on the plane $z = 0$; M_{0m} is the ellipticity of these waves; and $\kappa = 1/(1 + k_2/k_1)$. The origin of the polar coordinate system is located on the beam axis. The straight line $\varphi = 0$ is parallel to the principal axis of the polarisation ellipse of the wave $\boldsymbol{E}_1(\boldsymbol{r})$. The medium under study has the symmetry $\infty\infty$, so that the choice of this coordinate system does not violate the generality of the study. Expressions (11)–(14) were derived taking into account only the terms linear in small parameters $1/(k_1 w_1)$ and $1/(k_2 w_2)$ (in fact, in the divergence angles of pump beams).

3. Results and discussion

The dependence of the signal-wave intensity [expression (11)] on the polar angle φ for fixed values of r and z is rather simple:

$$I(r, \varphi, z) \sim \sigma_1 \cos 2\varphi + \sigma_2 \sin 2\varphi + \sigma_3, \qquad (15)$$

where $\sigma_{1,2,3} = \sigma_{1,2,3}(M_{01}, M_{02}, \Psi_2, \kappa)$. The maximum value of I is achieved on the straight lines $\varphi = \varphi_0$ and $\varphi = \varphi_0 + \pi$ ($0 \leqslant 2\Psi_2 < \pi/2$) or $\varphi = \varphi_0 + \pi/2$ and $\varphi = \varphi_0 + 3\pi/2$ ($\pi/2 < \Psi_2 < \pi$), while the minimum value is achieved on the straight lines $\varphi = \varphi_0 + \pi/2$ and $\varphi = \varphi_0 + 3\pi/2$ ($0 \leqslant \Psi_2 < \pi/2$) or $\varphi = \varphi_0$ and $\varphi = \varphi_0 + \pi$ ($\pi/2 \leqslant \Psi_2 < \pi$). In the latter expressions,

$$\varphi_0 = \frac{1}{2}\operatorname{arccot}\left(\cot 2\Psi_2 + \frac{R}{\sin 2\Psi_2}\right), \qquad (16)$$

where $R = (k_2/k_1)[(1 - M_{01}^2)/(1 - M_{02}^2)]^{1/2}$. Note that $\varphi_0 = \Psi_2$ for $R = 0$. If $\Psi_2 = \pi/2$, the maximum is achieved on the straight line $\varphi = 0$ ($R > 1$) or $\varphi = \pi/2$ ($R < 1$), and the minimum – on the straight line $\varphi = \pi/2$ ($R > 1$) or $\varphi = 0$ ($R < 1$). When both fundamental waves are circularly polarised or $R = 1$ and $\Psi_2 = \pi/2$, the intensity distribution is radially symmetric (I is independent of φ).

The roots of the equation $M(\varphi) = 0$ give polar angles determining in the beam cross section the directions of straight lines propagating through the beam centre on which radiation at the sum frequency is linearly polarised. For the nonnegative values of the parameter

$$D = M_{01}M_{02}\{[2\kappa(1-\kappa) - 1]M_{01}M_{02} + 2\kappa(1-\kappa)[\cos 2\Psi_2[(1 - M_{01}^2)(1 - M_{02}^2)]^{1/2} - 1]\} \qquad (17)$$

two such straight lines $\varphi = \varphi_+$ and $\varphi = \varphi_-$ exist, where

$$\varphi_\pm = \arctan\{[\kappa \sin(2\Psi_2)M_{01}(1 - M_{02}^2)^{1/2} \pm \sqrt{D}] \times \{\kappa M_{01}[(1 - M_{02}^2)^{1/2}\cos 2\Psi_2 - 1] + (1-\kappa)M_{02}[(1 - M_{02}^2)^{1/2} - 1]\}^{-1}\}. \qquad (18)$$

The beam is divided by them into four sectors so that the directions of rotation of the electric field vector $\boldsymbol{E}_\perp^{\rm sf}(r, \varphi)$ in neighbouring sectors are opposite.

The above-considered results are illustrated in Fig. 1a where the polarisation ellipses are shown at different points of the beam cross section at the sum frequency. The sum of squares of the half-axes of each of the ellipses is proportional to the radiation intensity at the ellipse centre (specified by the radius vector $\boldsymbol{r}$), the axial ratio of the ellipse is uniquely expressed in terms of $M(\boldsymbol{r})$, and the tilt angle of the principal axis coincides with the angle $\Psi(\boldsymbol{r})$. The direction of the electric field vector at a fixed instant at a point specified by the vector $\boldsymbol{r}$ is indicated with a small circle at the ellipse boundary. Light and dark ellipses correspond to the clockwise and counter-clockwise rotation of the vector $\boldsymbol{E}_\perp^{\rm sf}$, respectively. One can easily see that radiation at the sum frequency is polarised inhomogeneously. There exist regions of the beam with linear [$M(r, \varphi) = 0$], elliptic [$-1 < M(r, \varphi) < 1$], and circular [$M(r, \varphi) = \pm 1$] polarisations, and the direction of rotation of the electric field vector can change to the opposite with increasing φ. Also, variations in the intensity with increasing φ are observed (for fixed r), which are determined by expression (15), and a

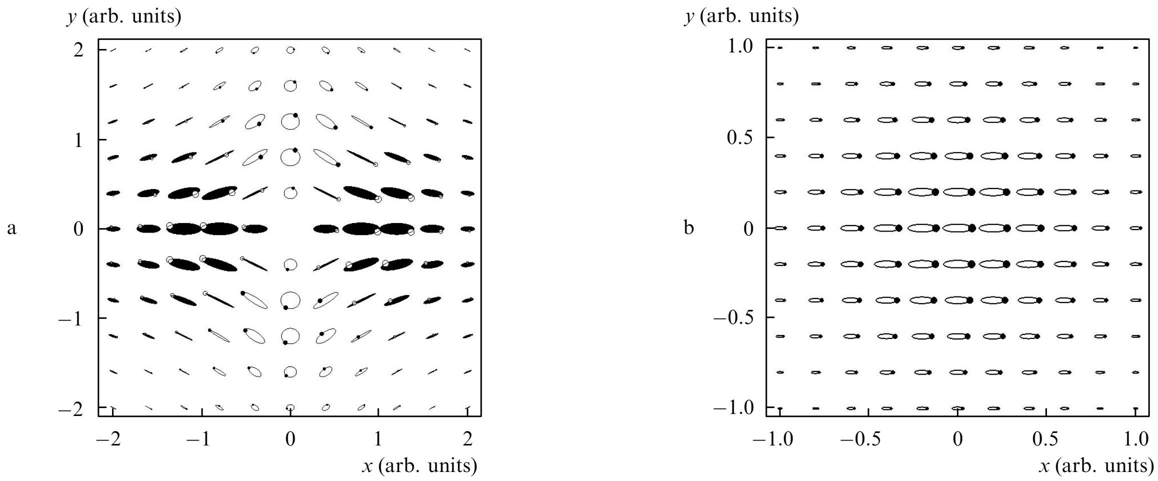

Figure 1. Transverse spatial distributions of polarisation of the sum-frequency beam for $M_{01} = 0.6$, $M_{02} = -0.4$, $\Psi_2 = \pi/2$, $k_2/k_1 = 2$ (a) and one of the fundamental beams for $M_{01} = 0.5$ (b).

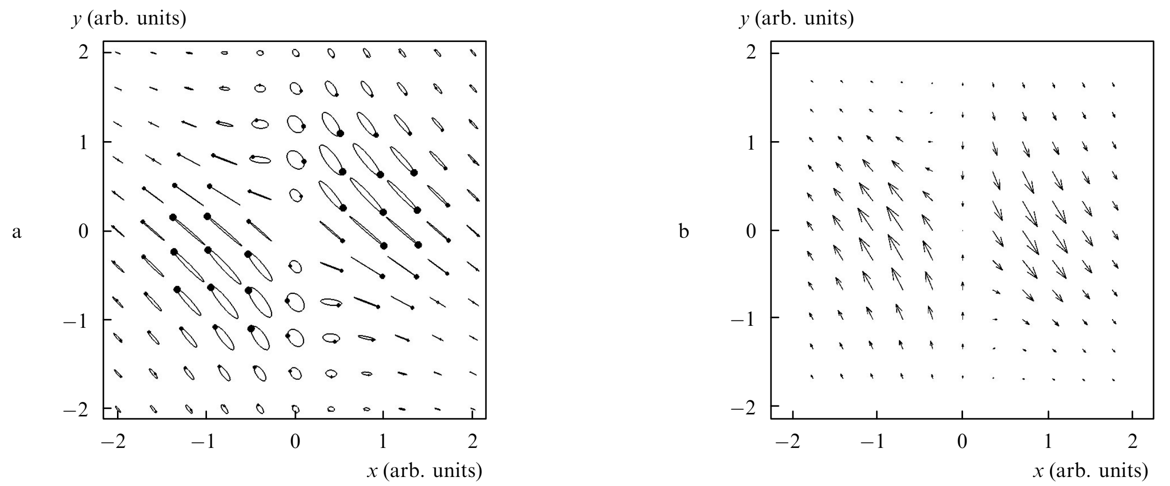

Figure 2. Transverse spatial distributions of polarisation of the sum-frequency beam for $M_{01} = 0.6$, $M_{02} = 0$, $\Psi_2 = \pi/3$, $k_2/k_1 = 2$ (a) and $M_{01} = 0$, $M_{02} = 0$, $\Psi_2 = \pi/4$, $k_2/k_1 = 2$ (b).

strong dependence of $\tilde{\alpha}$ on $\boldsymbol{r}$. For comparison, the transverse distribution of polarisation for one of the fundamental beams is shown in Fig. 1b. Unlike Fig. 1a, all the ellipses here are oriented identically and have the same eccentricity, and $\tilde{\alpha}(\boldsymbol{r}, z) \equiv 0$.

For $D = 0$, the straight lines $\varphi = \varphi_+$ and $\varphi = \varphi_-$ coincide and the direction of rotation of the electric-field vector in the beam cross section at the sum frequency does not change (Fig. 2a). If $M_{01} = 0$ and $M_{02} \neq 0$ ($M_{01} \neq 0$ and $M_{02} = 0$), the field $\boldsymbol{E}_{\perp}^{\rm sf}$ will have linear polarisation only on the straight line $\varphi = \pi/2$ ($\varphi = \pi/2 + \Psi_2$). The sum-frequency beam will be linearly polarised at all the points in the beam cross section only for $M_{01} = M_{02} = 0$; however, the direction of oscillations of the field $\boldsymbol{E}_{\perp}^{\rm sf}$ will coincide with the vector $\boldsymbol{r}$ only on the straight lines $\varphi = \pi/2$ и $\varphi = \pi/2 + \Psi_2$ (Fig. 2b).

Figure 3a shows polarisation ellipses at different points of the beam cross section at the sum frequency for $D < 0$. In this case, $M(\varphi) \neq 0$ and the direction of rotation of the electric field vector does not change. In the case of the oppositely directed circular polarisations of fundamental beams, the ellipticity at the sum frequency is independent of r and φ:

$$M(\varphi; M_{01} = \pm 1, M_{02} = \mp 1) = \pm 1 \mp \frac{(k_2/k_1)^2}{1 + (k_2/k_1)^2}, \quad (19)$$

and the principal axes of polarisation ellipses are oriented azimuthally (Fig. 3b).

Figure 4 shows the typical arrangement of the regions of ellipticity $M_{01,02}$ of fundamental waves, where the above-described variations in the polarisation of sum-frequency radiation occur, for different values of Ψ_2 and k_2/k_1. The light and grey regions correspond to $D > 0$ and $D < 0$, respectively. The thick lines correspond to $D = 0$. When M_{01} and M_{02} have the same signs, radiation is always elliptically polarised and the direction of rotation of the electric field vector does not change. The necessary condition for the appearance of regions with different polarisations (linear, elliptic, and circular) and opposite directions of rotation of the electric field vector is the negative sign of the product $M_{01}M_{02}$.

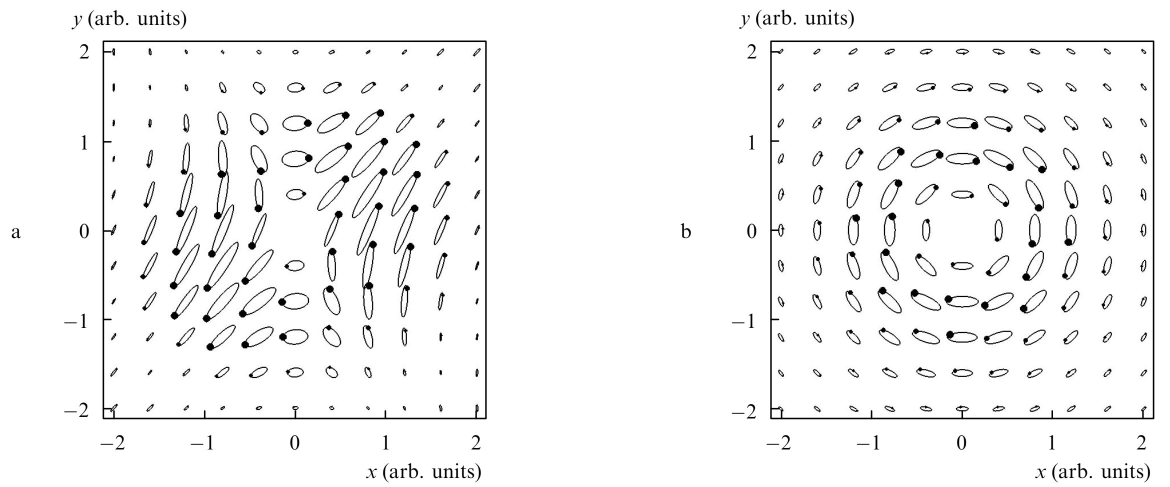

Figure 3. Transverse spatial distributions of polarisation of the sum-frequency beam for $M_{01} = 0.4$, $M_{02} = 0.2$, $\Psi_2 = 5\pi/12$, $k_2/k_1 = 2$ (a) and $M_{01} = 1$, $M_{02} = -1$, $k_2/k_1 = 2$ (b).

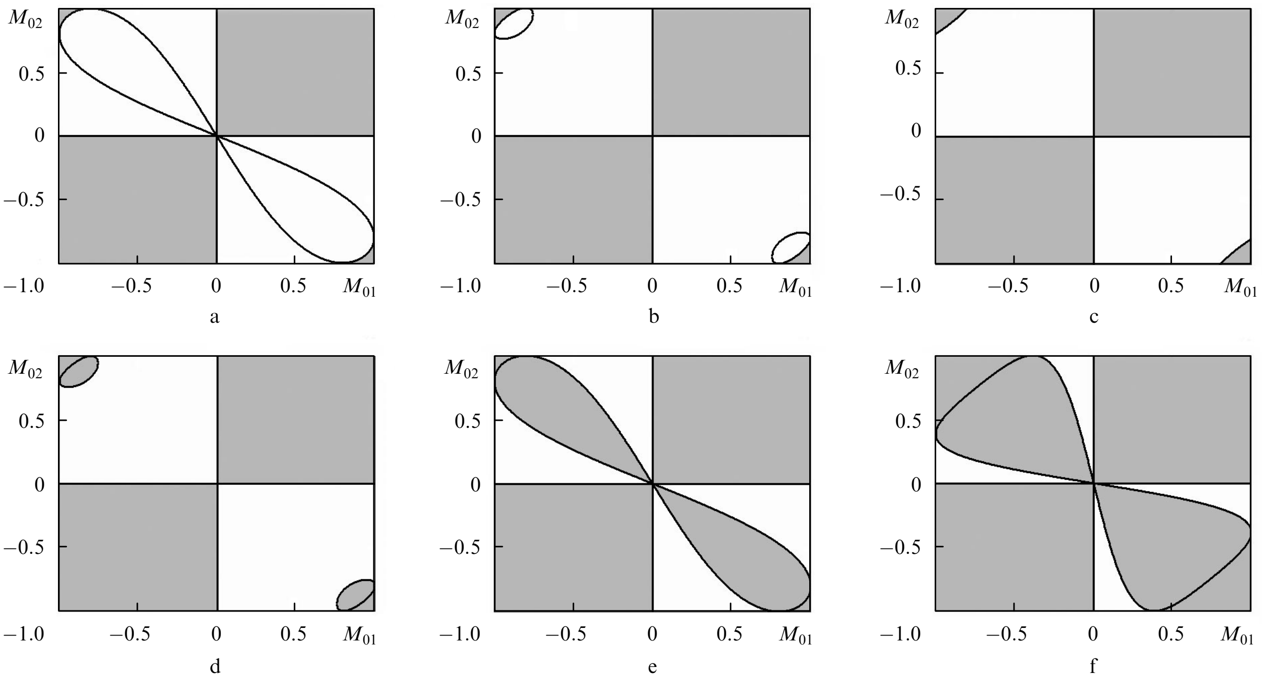

Figure 4. Regions corresponding to different regimes of sum-frequency generation in which $k_2/k_1 = 2$ and $\Psi_2 = \pi/2$ (a), $\pi/3$ (b), $\pi/4$ (c), and $\pi/6$ (d), and $\Psi_2 = 0$ and $k_2/k_1 = 2$ (e) and 5 (f).

4. Conclusions

Polarisation effects described above can be observed by the method proposed in [15] where the distribution of polarisation in the beam cross section was analysed upon the self-action of light. Radiation under study at frequency ω_{sf} is passed through a polariser and the transverse distribution of its intensity is recorded with a CCD array. The polariser is rotated with a small step and the intensity distribution is measured at each of its positions. The intensity values corresponding to different orientations of the polariser and measured by a pixel of the CCD array located at the point specified by the vector $\boldsymbol{r}$ allow us to measure $I(\boldsymbol{r})$, $M(\boldsymbol{r})$, and $\Psi(\boldsymbol{r})$, i.e., to determine the polarisation state of the light field at the point of the beam cross section where this pixel is located. The larger is the number of pixels of the CCD array, the better is the correspondence between the measured and real polarisation distributions. The appropriate dimensions of the beam incident on the CCD array can be obtained by using a system of lens.

Recall that the power of the sum-frequency signal and the dependence of the radiation intensity on the polar radius are determined not only by the polarisation of pump waves and the relation between their wave numbers specifying the change of polarisation in the beam cross section. The power also depends on the intensity and radii of the fundamental beams, the ratio of their diffraction lengths, the mismatch Δk of the wave vectors, beam-waist position in the medium and the medium length. The optimal choice of these parameters is discussed in detail in [7]. Under the most favourable conditions, the above-considered scheme for sum-frequency generation can be even more efficient than three-wave mixing in the noncollinear geometry. Calculations performed in [7] have shown that the coherent interaction length in the quasi-phase-matching region for the collinear geometry can achieve $(k_1 + k_2)w_1^2 w_2^2/[2(w_1^2 + w_2^2)]$, whereas in the case of a strongly noncollinear interaction this length is of the order of $1/(k_1 + k_2)$. Therefore, the efficiency increases approximately by a factor of $(k_1 + k_2)^2 w_1^2 w_2^2/(w_1^2 + w_2^2)$ due to the increase in the phase-matching length. An additional increase can be achieved by optimising all parameters of radiation and medium.

The results obtained in the paper have shown that the sum-frequency beam is always inhomogeneously polarised in its cross section (even in the simplest case when only the local quadratic susceptibility is taken into account and linearly polarised fundamental waves are considered in the collinear interaction geometry). This circumstance refutes the widespread opinion that polarisation changes weakly or insignificantly in nonlinear optical processes. Moreover, our preliminary studies have shown that drastic dependences of the intensity, parameters of the polarisation ellipse, and the orientation angle of the electric field vector at a fixed instant on transverse coordinates also appear in various problems of nonlinear optics. Among them are second harmonic generation from a chiral surface, self-focusing and compression of elliptically polarised light beams and pulses in an isotropic optically active medium. Despite this, at present the intensity of the signal wave rather than its polarisation is used in many known practical applications. The reason is the same mistake that polarisation does not change in nonlinear optical interactions. The results of our paper will undoubtedly stimulate the search for possible applications. Nevertheless, the information that the beam is inhomogeneous is very important for more exact calculations of its integrated parameters required to optimise the operation of quantum-electronic devices.

These results are also of interest for problems of nonlinear spectroscopy when it is necessary to analyse contributions from the components of tensors of local or nonlocal nonlinear susceptibilities to the process of interaction of waves. In this case, spectroscopic information is obtained by comparing the signal-wave intensities measured for different orientations of a polariser through which the wave passes (similarly to the measurements of the Stokes parameters). In particular, in experiments on sum-frequency generation (in the geometry considered in our paper), these intensities averaged over some region of the beam cross section will be measured. The average values of the intensity, ellipticity, and the rotation angle of the principal axis of the polarisation ellipse can be introduced by using different expressions with different weight factors in the integrand. When $I(\boldsymbol{r})$, $M(\boldsymbol{r})$, and $\Psi(\boldsymbol{r})$ described by expressions (11)–(13) change drastically, these average values can considerably change with increasing the size of the integration region. This circumstance should be taken into account especially in the case of local measurements of nonlinear susceptibilities, for example, in spatially inhomogeneous media and also measurements performed near the phase-transition temperature in liquid crystals, which can give unique information on the physics of the liquid-crystal state.

Our study has revealed the presence of the stringent relation between polarisations and wave numbers of the fundamental waves and the transverse structure of the sum-frequency beam field. This relation is found in our paper only for collinear homogeneosly polarised Gaussian beams propagating in a nonlinear isotropic gyrotropic medium. The obtained results are promising for the development of new methods for generating inhomogeneously polarised beams with the specified transverse structure drastically varying at relatively small scales. The latter is difficult to obtain by standard methods of linear optics. A separate interesting problem is the determination of optimal inhomogeneous distributions of the polarisation of fundamental waves allowing the generation of the signal wave at the sum frequency in a broad range of parameters with the homogeneous distribution of the required polarisation.

Acknowledgements. The authors thank S.A. Magnitskii for useful discussions and E.A. Khazanov for valuable critical comments.

References

1. Shen I.P. *Printsipy nelineinoi optiki* (Principles of Nonlinear Optics) (Moscow: Nauka, 1989).
2. Landsberg G.S. *Optika* (Optics) (Moscow: Nauka, 1976).
3. Makarov V.A. *Cand. Diss.* (Moscow State University, 1998).
4. Volkov S.N., Koroteev N.I., Makarov V.A. *Zh. Eksp. Teor. Fiz.*, **113**, 1261 (1998).
5. Volkov S.N., Koroteev N.I., Makarov V.A. *Opt. Spektr.*, **85**, 309 (1998).
6. Balakin A.V., Boucher D., Masselin P., Fertein E., Pakulev A.V., Resniansky A.Yu., Shkurinov A.P., Koroteev N.I. *Opt. Commun.*, **141**, 343 (1997).
7. Koroteev N.I., Makarov V.A., Volkov S.N. *Laser Phys.*, **9**, 655 (1998).
8. Rentzepis P.M., Giordmaine J.A., Wecht K.W. *Phys. Rev. Lett.*, **16**, 792 (1966).
9. Belkin M.A., Kulakov T.A., Ernst K.-H., et al. *Phys. Rev. Lett.*, **85**, 4474 (2000).
10. Belkin M.A., Han S.H., We X., Shen Y.R. *Phys. Rev. Lett.*, **87**, 113001-1 (2001).
11. Fischer P., Wiersma D.S., Righini R., et al. *Phys. Rev. Lett.*, **85**, 4253 (2000).
12. Champagne B., Fischer P., Buckingham A.D. *Chem. Phys. Lett.*, **331**, 83 (2000).
13. Belkin M.A., Kulakov T.A., Ernst K.-H., et al. *Opt. Mater.*, **21**, 1 (2002).
14. Fischer P., Beckwit K., Wis S.W., Albrech A.C. *Chem. Phys. Lett.*, **352**, 463 (2002).
15. Gahl A., Aumann A., Moeller M., Lange W. *J. Opt. B: Quantum Semiclass. Opt.*, **1**, 121 (1999)