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Nonuniform transverse distribution of the light intensity and polarisation upon sum-frequency generation from the surface of an isotropic gyrotropic medium in the case of normal incidence of light

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Abstract. **Sum-frequency generation from the surface of an isotropic gyrotropic medium by two normally incident homogeneously elliptically polarised Gaussian beams is studied theoretically. Analytic expressions, completely describing the transverse spatial distribution of the intensity and polarisation in the cross section of the reflected beam at the sum frequency, were derived taking into account both the local and nonlocal contributions of the quadratic nonlinearity of the medium thicknesses and the nonlinear contribution of its surface. It is shown that a special selection of the parameters of the fundamental waves allows one to determine components of the surface susceptibility tensor.**

Keywords: *sum-frequency generation, surface, elliptical polarisation, gyrotropy, spatial dispersion.*

1. Introduction

A rapid development of nonlinear optics, which opened to the world a variety of unusually beautiful physical phenomena, first overshadowed for some time the analysis of changes in polarisations of waves interacting in a medium (the history of the discovery and investigations of main nonlinear optical effects is presented in papers [1 – 3]). It was assumed initially that the states of their polarisations can produce only an insignificant influence on classical effects of nonlinear optics and, hence, a rather time-consuming theoretical investigation involving the solution of at least twice as many coupled nonlinear differential equations than in the approximation that the polarisation of light remains constant during light propagation, is hardly justified and is of academic interest only. Moreover, such studies were not stimulated experimentally. The estimates showed that some polarisation effects could be observed only at laser radiation intensities quite large for mid-1960s.

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Sergey Aleksandrovich Akhmanov never doubted the importance of constructing nonlinear polarisation optics. Together with V.I. Zharikov he predicted in 1967 the effect of nonlinear optical activity [4] – intensity-dependent rotation of the polarisation plane of linearly polarised light falling on a medium with a spatial dispersion of the cubic nonlinearity. This work stimulated subsequent development of nonlinear polarised optics at the Moscow State University (researches of N.I. Zheludev, V.A. Makarov and their disciples) and then at other scientific centres [2].

Theoretical and experimental investigations performed to date make it possible to assert definitely that the effects of polarisation self-action and interaction of waves belong to delicate but widespread effects of nonlinear optics. The use of the approximation of polarisation wave invariance during the propagation in theoretical calculations is hardly justified and represents only the first step to the subsequent description of nonlinear optical phenomena. A wave in quantum electronic devices is always elliptically polarised, the degree of its ellipticity and the inclination angle of the principal axis of the polarisation ellipse changing during the propagation through nonlinear crystals because of re-reflections from smooth surfaces and also because of resonator effects. Moreover, when waves interact in nonlinear media, their polarisation can change differently at different points of the light beam cross section [5 – 7]. In a number of cases, an elliptically polarised pulse can split into separate parts, the modulus of the degree of the electric field ellipticity in each part being close to unity. In this case, the rotation direction of the electric field vector in the pulse centre is opposite to the rotation direction in side parts [8].

The name of S.A. Akhmanov is also associated with the active application of ideas and methods of nonlinear optics in laser spectroscopic diagnostics of matter [9]. The extensive list of spectroscopic schemes proposed by Akhmanov and his disciples involves methods based on the use of intensity-dependent variations in light polarisation. Being one of the most advanced, the method for polarisation measurements [10] allows one to detect rather weak changes in the degree of ellipticity and the rotation angle of the principal axis of the polarisation ellipse of the signal wave and, hence, to obtain spectroscopic data on the matter, which are unavailable with the help of other investigation techniques. Additional possibilities are related to the use of elliptically polarised fundamental waves. The latter, according to S.A. Akhmanov, makes it possible to increase the number of 'degrees of freedom'. In other words, one can change the degree of ellipticity, the mutual orientation of the principal axes of polarisation ellipses and other parameters of the fundamental waves and, thereby to emphasise or suppress the contribution of local, nonlocal and (in problems related to the light reflection) surface nonlinear susceptibilities to the intensity and polarisation of a signal wave.

The sum-frequency generation (SFG), since the moment of discovery of this phenomenon, has been one of the most efficient and widely used investigation methods of surface properties. At present, the SFG is used in spectroscopy [11–13], microscopy [14–16], in excitation of surface waves [17] as well as in studying the order of molecules [18, 19] and molecular clusters on the medium surface or in a thin film deposited on a substrate. The SFG has been recently actively employed in problems related to the spectroscopy of vibrational spectra, thus stimulating the development of different models of radiation interaction with molecules [18, 20].

The SFG is an effective tool of spectroscopy and diagnostics of the surface of isotropic gyrotropic media. Unlike ordinary isotropic media (whose symmetry is $\infty\infty m$), these media consist of chaotically oriented chiral molecules, have the symmetry group $\infty\infty$, and, therefore, have a local quadratic susceptibility. In this case, the spatial dispersion of the quadratic response of the medium volume (related to the nonlocality of the medium response) and the quadratic nonlinearity of the surface also provide for the SFG. Note that the latter two SFG mechanisms will also take place in an ordinary isotropic medium.

In theoretical papers devoted to the SFG from the surface of an isotropic chiral medium, various attempts were made to take into account the effect of the spatial dispersion of the nonlinear optical response of the matter [21–28] and the inhomogeneity of the optical properties of the surface layer [26–28] as well as to show the means of the experimental separation of contributions to the signal wave, caused by the surface and the matter volume [26–28]. Calculations, as a rule, were performed in the plane-wave approximation and the wave vectors of interacting fundamental waves lay in one plane of incidence.

The authors of theoretical papers [29, 30] studied for the first time the influence of the spatial limitation of an incident elliptically polarised light beam on the second harmonic generation (SHG), which is a degenerate case of the SFG, from the surface of an isotropic gyrotropic medium. In [29], the specific properties of SHG were discussed in the case of the oblique incidence of a two-dimensional (slit) Gaussian beam, while the authors of [30] solved the SHG problem in the case of normal incidence of a three-dimensional Gaussian beam. In both papers special attention was paid to the accurate consideration of the nonlinear response of the matter surface (for this purpose modified boundary conditions were used [31, 32]) and the nonlocality of the nonlinear response of the medium volume.

It was shown in [29] that in the case of the oblique incidence of a two dimensional beam, its spatial limitation introduces only a small (proportional to the angle of its divergence) correction to the expression for the radiation field strength at the doubled frequency, which was obtained in the plane-wave approximation. It was established later [33] that in most cases the same can be said about a three-dimensional Gaussian beam falling at an arbitrary angle on a nonlinear medium with the parameters varying in a rather broad range. However, in the case of the normal incidence, the sum-frequency signal reflected from the surface of the isotropic chiral medium can emerge only due to the non-collinear interaction of spatial Fourier components of the beam [28, 30] (in the plane-wave approximation the SFG is impossible).

Spectroscopic schemes, in which the SHG is used in surface diagnostics in the case of normal incidence of elliptically polarised fundamental waves, allows one to obtain, with respect to the measured polarisation distribution in the plane of the transverse signal beam cross section, separate components of the tensors of nonlocal and surface nonlinear susceptibilities of the medium. This can be done [6] by fixing the polarisation states of radiation on specially selected straight lines in the plane of the reflected beam cross section at the doubled frequency. We can expect that in the case of the SFG it will be possible to acquire more data on the medium.

In this paper, we studied the specific character of the formation of an inhomogeneous polarisation distribution in the cross section of a sum-frequency beam appearing due to reflection of two normally incident coaxial elliptically polarised Gaussian fundamental beams from the surface of an isotropic gyrotropic medium (the $\infty\infty$ symmetry). We pay special attention to transverse polarisation distributions, which make it possible to extract spectroscopic information on the chiral medium or its surface.

2. Method for finding the electric field strength in the reflected sum-frequency wave and solution of the problem in quadratures

A reflected beam at the sum frequency $\omega_3 = \omega_1 + \omega_2$ emerges due to nonlinear optical responses of the surface of a medium and its volume caused by monochromatic waves at frequencies $\omega_{1,2}$. The first of them is related to the difference in the symmetry of the surface layer of the isotropic gyrotropic medium (group ∞) from the symmetry of its thickness (group $\infty\infty$). We will describe it by using the modified boundary conditions for the electromagnetic field [31]. They are obtained by solving Maxwell's equations in the surface layer of the medium with the effective thickness d_0.

Let us couple the coordinate system with the surface of the medium under study so that the z axis be directed perpendicular to the surface inside the medium and axes x and y be on the surface. In the first approximation in the small parameter $\mu \simeq d_0/\lambda_m$, where $\lambda_m = 2\pi c/\omega_m$ and $m = 1, 2, 3$, these conditions relating the vector components of the strength $\boldsymbol{E}$ and inductions $\boldsymbol{D}$ и $\boldsymbol{B}$ of the electric and magnetic fields at the frequency ω_m in vacuum (the superscript 'v') and in matter (the superscript 't') with the nonlocality of the nonlinear optical response have the form at the interface $z = 0$ [31]:

$$\boldsymbol{E}^{(\mathrm{v})}_{\tan}(\omega_m) - \boldsymbol{E}^{(\mathrm{t})}_{\tan}(\omega_m) = \frac{4\pi}{\mathrm{i}\omega_m}\mathrm{grad}_{\tan} i_{\mathrm{n}}(\omega_m),$$

$$D^{(\mathrm{v})}_{\mathrm{n}}(\omega_m) - D^{(\mathrm{t})}_{\mathrm{n}}(\omega_m) = \frac{4\pi}{\mathrm{i}\omega_m}\mathrm{div}\,\boldsymbol{i}_{\tan}(\omega_m), \tag{1}$$

$$\boldsymbol{B}^{(\mathrm{v})}(\omega_m) - \boldsymbol{B}^{(\mathrm{t})}(\omega_m) = \frac{4\pi}{c}[\boldsymbol{n} \times \boldsymbol{i}_{\tan}(\omega_{\boldsymbol{m}})].$$

Here, $\boldsymbol{n}$ is the surface-perpendicular unit vector directed opposite to the z axis from the medium to the vacuum; c is the velocity of light; subscripts 'tan' and 'n' denote tangential and normal components of the vectors $\boldsymbol{E}$, $\boldsymbol{D}$, $\boldsymbol{B}$ и $\boldsymbol{i}$. The latter can be interpreted as a surface current density of coupled charges [31, 32] and in the general case in the first approximation in the parameter μ it can be represented in the form of expansion in powers $\boldsymbol{E}^{(\mathrm{v})}$:

$$\boldsymbol{i} = \hat{\kappa}^{(1)}\boldsymbol{E}^{(\mathrm{v})} + \hat{\kappa}^{(2)} : \boldsymbol{E}^{(\mathrm{v})}\boldsymbol{E}^{(\mathrm{v})} + \hat{\kappa}^{(3)} \vdots\ \boldsymbol{E}^{(\mathrm{v})}\boldsymbol{E}^{(\mathrm{v})}\boldsymbol{E}^{(\mathrm{v})} + \ldots, \quad (2)$$

where the material tensors $\hat{\kappa}^{(n)}$ characterise the surface response of the nonlinear medium to external electromagnetic field. The reflected sum-frequency signal caused by the thin surface layer is related to the second term in this sum:

$$i_i(\omega_3) = \kappa^{(2)}_{ikl}(\omega_3;\omega_1,\omega_2)E^{(\mathrm{v})}_k(\omega_1)E^{(\mathrm{v})}_l(\omega_2). \quad (3)$$

To find the nonlinear optical response of the medium thickness caused by monochromatic waves at frequencies $\omega_{1,2}$, we will use the phenomenological approach and will write the expression for the nonlinear polarisation at the frequency ω_3 in the form:

$$P^{\mathrm{NL}}_i(\omega_3) = \chi^{(2)}_{ijk}(\omega_3;\omega_1,\omega_2)E^{(\mathrm{t})}_j(\omega_1)E^{(\mathrm{t})}_k(\omega_2) + \gamma^{(2)1}_{ijkl}(\omega_3;\omega_1,\omega_2)\frac{\partial E^{(\mathrm{t})}_k(\omega_1)}{\partial x_j}E^{(\mathrm{t})}_l(\omega_2) + \gamma^{(2)2}_{ijkl}(\omega_3;\omega_1,\omega_2)E^{(\mathrm{t})}_k(\omega_1)\frac{\partial E^{(\mathrm{t})}_l(\omega_2)}{\partial x_j}. \quad (4)$$

Here, $\hat{\chi}^{(2)}$ is the tensor of the local quadratic susceptibility of the medium volume; $\hat{\gamma}^{(2)}$ is the tensor describing weak spatial dispersion appearing due to the nonlocality of the quadratic response of the medium volume. The consideration of the spatial dispersion and determination of the material tensor $\hat{\gamma}^{(2)}$ describing it at a small (compared to the length of the propagating wave) scale of the optical response nonlocality is described in detail in [27–30]. Note that, unlike $\hat{\chi}^{(2)}$, tensors $\hat{\gamma}^{(2)1,2}$ describing the spatial dispersion of the quadratic nonlinearity are nonsymmetric with respect to the permutation of two last subscripts with a simultaneous permutation of frequency arguments.

Consider weakly diverging monochromatic fundamental beams (having the common symmetry axis, which coincides with the z axis) with arbitrary intensity and polarisation distributions, which fall collinearly on the surface of the isotropic gyrotropic medium. By assuming that the characteristic changes in the electric field strength in the cross section planes of incident, reflected and refracted beams occur at distances much larger than λ_m, we will represent $\boldsymbol{E}^{(\mathrm{v,t})}(\omega_m, \boldsymbol{r}, z=0)$, by using the Fourier integral, in the form of superposition of monochromatic plane waves:

$$\boldsymbol{E}^{(\mathrm{v,t})}(\omega_m,\boldsymbol{r},z=0) = \iint \tilde{\boldsymbol{E}}^{(\mathrm{v,t})}(\omega_m,\boldsymbol{k}_{m\perp})\exp(\mathrm{i}\boldsymbol{k}_{m\perp}\boldsymbol{r})\mathrm{d}\boldsymbol{k}_{m\perp}, \quad (5)$$

where $\boldsymbol{k}_{m\perp}$ is the component of the wave vector lying in the xy plane. We will assume below that absorption is absent in the medium and all the spatial Fourier components $\tilde{\boldsymbol{E}}^{(\mathrm{t})}(\omega_m,\boldsymbol{k}_{1,2\perp})$ have real wave vectors. Taking into account the smallness of the angular divergence of the fundamental beams, in determining the field we will allow only for linear terms in $\boldsymbol{k}_{1,2,3\perp}$ and in exponents (where they are absent) – quadratic terms. $\tilde{\boldsymbol{E}}^{(\mathrm{v})}(\omega_{1,2},\boldsymbol{k}_{1,2\perp})$ and $\tilde{\boldsymbol{E}}^{(\mathrm{t})}(\omega_{1,2},\boldsymbol{k}_{1,2\perp})$ entering (5) can be readily expressed via Fourier transforms of the fields of incident waves $\tilde{\boldsymbol{E}}_{\perp}(\omega_{1,2},\boldsymbol{k}_{1,2\perp})$ by setting $\boldsymbol{i}(\omega_{1,2}) \equiv 0$ in (1) [in this case, conditions (1) become conventional boundary conditions] and passing in them to spatial Fourier components. As a result, by neglecting $P^{\mathrm{NL}}_i(\omega_{1,2})$, we obtain the relations:

$$\tilde{\boldsymbol{E}}^{(\mathrm{v})}_{\perp}(\omega_{1,2},\boldsymbol{k}_{1,2\perp}) = \tilde{\boldsymbol{E}}^{(\mathrm{t})}_{\perp}(\omega_{1,2},\boldsymbol{k}_{1,2\perp}) = \frac{2}{1+n_{1,2}}\tilde{\boldsymbol{E}}_{\perp}(\omega_{1,2},\boldsymbol{k}_{1,2\perp}), \quad (6)$$

$$\tilde{E}^{(\mathrm{v})}_z(\omega_{1,2},\boldsymbol{k}_{1,2\perp}) = \varepsilon_{1,2}\tilde{E}^{(\mathrm{t})}_z(\omega_{1,2},\boldsymbol{k}_{1,2\perp}) = -\varepsilon_{1,2}\frac{2}{(1+n_{1,2})n_{1,2}k_{1,2}}\boldsymbol{k}_{1,2\perp}\tilde{\boldsymbol{E}}_{\perp}(\omega_{1,2},\boldsymbol{k}_{1,2\perp}), \quad (7)$$

where $\varepsilon_{1,2}$ is the dielectric constant of the medium at frequencies $\omega_{1,2}$ respectively; $n_{1,2} = \sqrt{\varepsilon_{1,2}}$ are the refractive indices; $\tilde{\boldsymbol{E}}(\omega_{1,2},\boldsymbol{k}_{1,2\perp})$ are spatial Fourier components of the electric fields of fundamental beams falling on the medium. Recall that $\tilde{\boldsymbol{E}}^{(\mathrm{v})}(\omega_{1,2},\boldsymbol{k}_{1,2\perp}) = \tilde{\boldsymbol{E}}(\omega_{1,2},\boldsymbol{k}_{1,2\perp}) + \tilde{\boldsymbol{E}}^{(\mathrm{r})}(\omega_{1,2},\boldsymbol{k}_{1,2\perp})$, where the superscript 'r' corresponds to the reflected wave. Knowing the latter and representing polarisation $\boldsymbol{P}^{\mathrm{NL}}_i(\omega_3)$ and current $\boldsymbol{i}(\omega_3)$ similarly to (5), in the form of Fourier integrals and also taking into account that the wave vector component lying in the xy plane is $\boldsymbol{k}_{3\perp} = \boldsymbol{k}_{1\perp} + \boldsymbol{k}_{2\perp}$, we can find the expressions for the spatial Fourier components of polarisation $\tilde{\boldsymbol{P}}^{(\mathrm{NL})}(\omega_3,\boldsymbol{k}_{3\perp})$ and current $\tilde{\boldsymbol{i}}(\omega_3,\boldsymbol{k}_{3\perp})$:

$$\tilde{i}_j(\omega_3,\boldsymbol{k}_{3\perp}) = \iint \kappa^{(2)}_{jkl}(\omega_3;\omega_1,\omega_2) \times \tilde{E}^{(\mathrm{v})}_k(\omega_1,\boldsymbol{k}_{1\perp})\tilde{E}^{(\mathrm{v})}_l(\omega_2,\boldsymbol{k}_{3\perp}-\boldsymbol{k}_{1\perp})\mathrm{d}\boldsymbol{k}_{1\perp}, \quad (8)$$

$$\begin{aligned}\tilde{P}^{(\mathrm{NL})}_i(\omega_3,\boldsymbol{k}_{3\perp}) &= \chi^{(2)}_{ijk}(\omega_3;\omega_1,\omega_2) \times \iint \tilde{E}^{(\mathrm{t})}_j(\omega_1,\boldsymbol{k}_{1\perp})\tilde{E}^{(\mathrm{t})}_k(\omega_2,\boldsymbol{k}_{3\perp}-\boldsymbol{k}_{1\perp})\mathrm{d}\boldsymbol{k}_{1\perp} \\ &+ \gamma^{(2)1}_{ijkl}(\omega_3;\omega_1,\omega_2)\iint k_{1j}\tilde{E}^{(\mathrm{t})}_k(\omega_1,\boldsymbol{k}_{1\perp}) \times \tilde{E}^{(\mathrm{t})}_l(\omega_2,\boldsymbol{k}_{3\perp}-\boldsymbol{k}_{1\perp})\mathrm{d}\boldsymbol{k}_{1\perp} + \gamma^{(2)2}_{ijkl}(\omega_3;\omega_1,\omega_2) \\ &\times \iint (k_{1j}-k_{3j})\tilde{E}^{(\mathrm{t})}_k(\omega_1,\boldsymbol{k}_{1\perp})\tilde{E}^{(\mathrm{t})}_l(\omega_2,\boldsymbol{k}_{3\perp}-\boldsymbol{k}_{1\perp})\mathrm{d}\boldsymbol{k}_{1\perp}.\end{aligned} \quad (9)$$

One can see that expressions (8) and (9) represent convolutions of spatial Fourier field components of the waves incident on the medium, which are calculated by integration with respect to all possible values of $\boldsymbol{k}_{1\perp}$ and $\boldsymbol{k}_{2\perp}$ (their sum is equal to $\boldsymbol{k}_{3\perp}$).

To calculate the spatial Fourier components of the electric field strength of the reflected wave at the sum frequency $\tilde{\boldsymbol{E}}^{(\mathrm{r})}(\omega_3,\boldsymbol{k}_{3\perp})$, it is necessary to substitute into modified boundary conditions (1) spatial Fourier components (8) of the surface current density of coupled charges $\tilde{\boldsymbol{i}}(\omega_3,\boldsymbol{k}_{3\perp})$ and the electric field induction

$$\tilde{\boldsymbol{D}}^{(t)}(\omega_3, \boldsymbol{k}_{3\perp}) = \varepsilon_3 \tilde{\boldsymbol{E}}^{(t)}(\omega_3, \boldsymbol{k}_{3\perp}) + 4\pi \tilde{\boldsymbol{P}}^{(NL)}(\omega_3, \boldsymbol{k}_{3\perp}), \quad (10)$$

satisfying the equation $\mathrm{div}\tilde{\boldsymbol{D}}^{(t)}(\omega_3, \boldsymbol{k}_{3\perp}) = 0$ and then to solve the derived system of algebraic equations. In (10), ε_3 is the dielectric constant of the medium at the frequency ω_3. To find $\tilde{\boldsymbol{D}}^{(t)}(\omega_3, \boldsymbol{k}_{3\perp})$, it is necessary to know the spatial Fourier components $\tilde{\boldsymbol{E}}^{(t)}(\omega_3, \boldsymbol{k}_{3\perp})$, which can be determined by solving in the fixed-field approximation the wave equation with nonlinear polarisation (9) in the right-hand side. As a result, we obtain expressions for spatial Fourier components constituting the electric field strengths of the reflected sum-frequency wave near the medium surface:

$$\tilde{\boldsymbol{E}}_{\perp}^{(r)}(\boldsymbol{k}_{3\perp}) = -\frac{4\pi}{n_3}\left[\frac{1}{c}\left(\tilde{\boldsymbol{i}}_{\perp} + \frac{n_3 \boldsymbol{k}_{3\perp}}{k_3^{(r)}}\tilde{i}_z\right) + \frac{\omega_3}{n_1\omega_1 + n_2\omega_2 + n_3\omega_3}\left(\tilde{\boldsymbol{P}}_{\perp}^{(NL)} + \frac{\boldsymbol{k}_{3\perp}}{n_3 k_3^{(r)}}\tilde{P}_z^{NL}\right)\right], \quad (11)$$

$$\tilde{E}_z^{(r)}(\omega_3, \boldsymbol{k}_{3\perp}) = \frac{1}{k_3^{(r)}}(\boldsymbol{k}_{3\perp}\tilde{\boldsymbol{E}}_{\perp}^{(r)}(\omega_3, \boldsymbol{k}_{3\perp})), \quad (12)$$

where $n_3 = \sqrt{\varepsilon_3}$, a $k_3^{(r)} = \omega_3/c$. One can see from the expressions derived that the longitudinal component $\tilde{E}_z^{(r)}(\omega_3, \boldsymbol{k}_{3\perp})$ has a higher order of smallness with respect to the angular divergence of the beam than the transverse components $\tilde{\boldsymbol{E}}_{\perp}^{(r)}(\boldsymbol{k}_{3\perp})$. By substituting $\tilde{\boldsymbol{E}}_{\perp}^{(r)}(\boldsymbol{k}_{3\perp})$ and $\tilde{E}_z^{(r)}(\omega_3, \boldsymbol{k}_{3\perp})$ into (5) and performing integration, we can find in quadratures the intensity and polarisation distribution in the plane of the reflected beam cross section at the sum frequency near the medium surface. It is pertinent to note that it can be done for any distributions of the electric field strengths of the fundamental beams with a small angular divergence.

3. Case of incidence of homogeneously elliptically polarised Gaussian beams on the medium

Consider two medium-incident fundamental beams with the Gaussian intensity profile

$$\boldsymbol{E}(\omega_{1,2}, x, y, z) = \left[\boldsymbol{e}_{1,2} + \frac{\mathrm{i}}{k_{1,2}^{(r)}}\boldsymbol{e}_z(\boldsymbol{e}_{1,2}\nabla)\right] \times \frac{E_{01,02}}{\beta_{1,2}(z)}\exp\left(-\frac{x^2+y^2}{w_{1,2}^2\beta_{1,2}(z)} - \mathrm{i}\omega_{1,2}t + \mathrm{i}k_{1,2}z\right). \quad (13)$$

which are homogeneously elliptically polarised in the plane of the cross section. Here, $|\boldsymbol{e}_{1,2}|^2 = 1$ are complex unit vectors of polarisation (their concrete form will be determined below); $\beta_{1,2}(z) = 1 - \mathrm{i}z/l_{1,2}$; $l_{1,2} = k_{1,2}w_{1,2}^2/2$ are diffraction lengths; $E_{01,02}$ are the amplitudes, $\omega_{1,2}$ are the frequencies, $w_{1,2}$ are the half-widths of the beams of fundamental radiation; $k_{1,2} = \omega_{1,2}/c$ are the corresponding wave numbers. Expression (13) contains a longitudinal component of the electric field strength in the form for which in the first approximation in the parameter $\lambda_{1,2}/w_{1,2}$ (the angles of divergence of incident beams are assumed small, $w_{1,2} \gg \lambda_{1,2}$) in vacuum the necessary condition

$$\mathrm{div}\boldsymbol{E}(\omega_{1,2}) = 0 \quad (14)$$

is fulfilled. Note that the planes of beam waists given by expression (13) coincide with the plane $z = 0$.

Elliptically polarised radiation is completely characterised by two complex or four real parameters. One can select circularly polarised field components $E_{m\pm} = E_x(\omega_m) \pm \mathrm{i}E_y(\omega_m)$ or Stokes parameters. However, in our problem it is convenient to use the following four parameters: the intensity $I_m = (|E_{m+}|^2 + |E_{m-}|^2)/2$, the degree of ellipticity $M_m = (|E_{m+}|^2 - |E_{m-}|^2)/(|E_{m+}|^2 + |E_{m-}|^2)$, the rotation angle of the principal axis of the polarisation ellipse $\Psi_m = \frac{1}{2}\arg\{E_{m+}E_{m-}^*\}$ and the angle specifying the orientation of the electric field vector at a fixed instant of time, $\Phi_m = \arg\{E_{m+} + E_{m-}^*\}$. In homogeneously polarised beams, M, Φ and Ψ are independent of transverse coordinates. Recall that M changes from -1 (left-hand circular polarisation) to $+1$ (right-hand circular polarisation) by passing through zero (linear polarisation), while Ψ – from 0 to π (the states 0 and π are equivalent).

It is easy to show that in this case, unit vectors $\boldsymbol{e}_m$ entering (13), without the loss of generality and assuming that $\Phi(\omega_{1,2}) = 0$, can be written in the form:

$$\boldsymbol{e}_m = [(1 - M_{0m})^{1/2}\exp(-\mathrm{i}\Psi_m)\boldsymbol{e}_+ + (1 + M_{0m})^{1/2}\exp(\mathrm{i}\Psi_m)\boldsymbol{e}_-]/\sqrt{2}, \quad (15)$$

where $|\boldsymbol{e}_+|^2 = |\boldsymbol{e}_-|^2 = 1$ and $(\boldsymbol{e}_+\boldsymbol{e}_-^*) = 0$; M_{01} and M_{02} are the degrees of ellipticity of radiation in homogeneously polarised beams incident on the medium; the angles Ψ_1 and Ψ_2 are given by the orientation of the principal axes of their polarisation ellipses.

After calculating, using (13), the spatial Fourier components of the electric field strengths of incident beams and deriving with their help expressions for $\tilde{\boldsymbol{i}}$ and $\tilde{\boldsymbol{P}}^{(NL)}$, we will write the expression for $\tilde{\boldsymbol{E}}^{(r)}(\boldsymbol{k}_{3\perp})$ by using (11) and (12). After integrating it in all possible values of $\boldsymbol{k}_{3\perp}$, we will find the field of the reflected beam at the frequency ω_3 in the form:

$$\begin{aligned} E_{\pm}^{(r)}(\omega_3, r, \varphi, z) = -D(r,z)\{&\exp(\pm\mathrm{i}\varphi)[(C_{0\pm} - C_{2\pm})\times \\ &\times[(1 \pm M_{01})(1 \mp M_{02})]^{1/2}\exp[\pm\mathrm{i}(\Psi_1 - \Psi_2)] \\ &+ (C_{0\mp} - C_{1\pm})[(1 \mp M_{01})(1 \pm M_{02})]^{1/2}\exp[\mp\mathrm{i}(\Psi_1 - \Psi_2)]] \\ &- \exp(\mp\mathrm{i}\varphi)(C_{1\pm} + C_{2\pm})[(1 \pm M_{01})(1 \pm M_{02})]^{1/2} \\ &\times\exp[\pm\mathrm{i}(\Psi_1 + \Psi_2)]\}, \end{aligned} \quad (16)$$

where $x = r\cos\varphi$; $y = r\sin\varphi$; r and φ are the polar radius and angle;

$$D(r,z) = \frac{8\sqrt{2}\pi\mathrm{i}E_{01}E_{02}}{(1+n_1)(1+n_2)(1+n_3)\omega_3 w_3^2}\frac{r}{\beta_3^2(z)} \times\exp\left[-\frac{r^2}{w_3^2\beta_3(z)} - \mathrm{i}(k_3 z + \omega_3 t)\right]; \quad (17)$$

$w_3^2 = w_1^2 w_2^2/(w_1^2 + w_2^2)$; $\beta_3(z) = 1 - \mathrm{i}z/l_3$; $l_3 = k_3 w_3^2/2$; $k_3 = \omega_3/c$. The coefficients $C_{0\pm}$, $C_{1\pm}$ and $C_{2\pm}$ in expression (16) depend on the medium parameters, on ω_1/ω_2, w_1/w_2 and have the form

$$C_{0\pm} = n_3 b_1 \pm \mathrm{i}(n_3 b_7 + \xi_\omega\chi/n_3) + \mathrm{i}\xi_\omega[(w_2^2\gamma_1 + w_1^2\gamma_2)$$

$$\times\,(w_1^2+w_2^2)^{-1}+\,(n_1\omega_1\gamma_1+n_2\omega_2\gamma_2)/n_3\omega_3],$$

$$C_{1\pm}=(l_3/l_1)\{n_1b_4\pm\mathrm{i}(n_1b_6+\xi_\omega\chi/n_1)+\mathrm{i}\xi_\omega\gamma_4$$
$$\times\,[(n_2l_2-n_1l_1)/n_2l_2](n_2\omega_2/n_1\omega_3)\},\tag{18}$$

$$C_{2\pm}=(l_3/l_2)\{n_2b_3\mp\mathrm{i}(n_2b_5+\xi_\omega\chi/n_2)-\,\mathrm{i}\xi_\omega\gamma_3$$
$$\times\,[(n_2l_2-n_1l_1)/n_1l_1](n_1\omega_1/n_2\omega_3)\}.$$

Here, $\xi_\omega=\omega_3/(n_1\omega_1+n_2\omega_2+n_3\omega_3)$; χ is the constant determining all nonzero tensor components of the quadratic response of the medium thickness $\chi_{ijk}^{(2)}=(\chi/c)e_{ijk}$; e_{ijk} is the Levi – Civita symbol. Expression (18) contains six out of seven independent tensor components of the quadratic response of the medium surface $\hat{\kappa}^{(2)}(\omega_3)$: $b_1=\kappa_{zxx}^{(2)}=\kappa_{zyy}^{(2)}$, $b_2=\kappa_{zzz}^{(2)}$, $b_3=\kappa_{yyz}^{(2)}=\kappa_{xxz}^{(2)}$, $b_4=\kappa_{xzx}^{(2)}=\kappa_{yzy}^{(2)}$, $b_5=\kappa_{xyz}^{(2)}=-\kappa_{yxz}^{(2)}$, $b_6=\kappa_{yzx}^{(2)}=-\kappa_{xzy}^{(2)}$, and $b_7=\kappa_{zxy}^{(2)}=-\kappa_{zyx}^{(2)}$. These components are specified in the crystal-physical coordinate system coinciding with xyz (because the medium itself and its surface layer are isotropic, an arbitrary choice of directions of x and y axes is possible). The same expression includes tensor components multiplied by ω_3, which characterise the spatial dispersion of the quadratic optical response of the medium volume: $\gamma_{1,2}=\omega_3\gamma_{xxyy}^{(2)1,2}$, $\gamma_3=\omega_3\gamma_{xyxy}^{(2)1}$ and $\gamma_4=\omega_3\gamma_{xyyx}^{(2)2}$. Let us emphasise that expressions (16) – (18) also contain terms found in the first approximation with respect to the parameter λ_m/w_m.

If $\hat{\gamma}^{(2)1}=\hat{\gamma}^{(2)2}=0$, then $C_{0,1,2+}=C_{0,1,2-}^{*}$. In this case, if the polarisation of incident waves is linear, the sum-frequency signal is also linearly polarised at all the points of the reflected beam cross section.

In the case of equal degrees of ellipticity of incident beams and $n_1l_1=n_2l_2$, expression (16) is considerably simplified. It is easy to notice that in this case the sum-frequency signal is independent of the tensor $\hat{\chi}^{(2)}$ at equal orientations of principal axes of polarisation ellipses of incident waves Ψ_1 and Ψ_2. If $|\Psi_1-\Psi_2|=\pi/2$, i.e. the principal axes of these ellipses are mutually perpendicular, the sum-frequency field is independent of the tensor components $\hat{\gamma}^{(2)1,2}$.

The latter circumstance allows one to distinguish the response of the medium surface from the response of its volume. For example, if $\hat{\chi}^{(2)}\equiv 0$, due to the presence of the symmetry centre, the absence of dispersion in the frequency rage employed or other reasons, the choice of Ψ_1 and Ψ_2, satisfying the condition $|\Psi_1-\Psi_2|=\pi/2$, guarantees the complete absence of the contribution of the medium volume to reflected radiation at the sum frequency.

Expression (16) allows one to calculate the intensity $I_3(r,\varphi,z)$, the degree of ellipticity $M_3(r,\varphi,z)$ and the rotation angle of the principal axes of the polarisation ellipse $\Psi_3(r,\varphi,z)$ of radiation at the sum frequency. If the beams incident on the medium are circularly polarised in opposite directions ($M_{01}=-M_{02}=\pm1$), then $M_3=\tilde{M}_3$, where the constant

$$\tilde{M}_3=\pm|C_{2\pm}|^2\mp|C_{1\mp}|^2\pm(C_{1\mp}C_{0\pm}^{*}+\text{c.c.})$$
$$\mp(C_{2\pm}C_{0\pm}^{*}+\text{c.c.})[2|\mathrm{C}_{0\pm}|^2+|\mathrm{C}_{2\pm}|^2+|\mathrm{C}_{1\mp}|^2$$
$$-(C_{1\mp}C_{0\pm}^{*}+\text{c.c.})-(C_{2\pm}C_{0\pm}^{*}+\text{c.c.})]^{-1},\tag{19}$$

and the rotation angle of the principal axis of the polarisation ellipse Ψ_3 differs from φ by the constant:

$$\Psi_3=\varphi\pm0.5\arg(|\mathrm{C}_{0\pm}|^2-\mathrm{C}_{2\pm}\mathrm{C}_{0\pm}^{*}$$
$$-C_{0\pm}C_{1\mp}^{*}+C_{2\pm}C_{1\mp}^{*}).\tag{20}$$

Figure 1 illustrates this case when each ellipse contains information on the electromagnetic field at the point of the beam cross section corresponding to the centre of the depicted ellipse. The radiation intensity in it is proportional to the sum of squares of lengths of its semiaxes, and M_3 and Ψ_3 coincide with analogous parameters of the ellipse in the figure. The points at its boundary indicate the direction of the electric field vector at the ellipse centre at the instant $t=k_3^{(\mathrm{r})}z_1/\omega_3$. In the case depicted in Fig. 1, the electric field vector rotates counterclockwise along the ellipses.

The electromagnetic field distribution in Fig. 1 can change rather strongly with varying ω_1/ω_2 and w_1/w_2, which is shown in Fig. 2. This figure presents the dependences of $\tilde{M}_3$ on ω_1/ω_3 and w_1/w_2. One can see that with increasing ω_1/ω_3, $\tilde{M}_3$ can change from -1 to $+1$. If $\hat{\kappa}^{(2)}(\omega_3)\equiv0$ and $n_1l_1=n_2l_2$, $\tilde{M}_3=0$, and $\Psi_3=\varphi$. In this case, the sum-frequency beam is radially polarised (Fig. 3). This distribution of the intensity and polarisation appears in some other particular cases.

Analysis of the dependence $M_3(\varphi)$ shows that the equation $M_3(\varphi)=0$ is quadratic with respect to $\tan\varphi$. If $\Psi_1=\Psi_2=0$ and $M_{01}=M_{02}=0$, its first root is $\varphi_{01}=\pi/2$, and the second root φ_{02}, is given by the expression

$$\varphi_{02}=-\arctan\{[((w_2^2\gamma_1+w_1^2\gamma_2)/(w_1^2+w_2^2)$$
$$+(n_1\omega_1\gamma_1+n_2\omega_2\gamma_2)/n_3\omega_2+l_3(n_2l_2-n_1l_1)\times$$
$$\times\,(n_1\omega_1\gamma_3-n_2\omega_2\gamma_4)/n_1n_2l_1l_2\omega_3)(n_2b_5/l_2-n_1b_6/l_1+$$

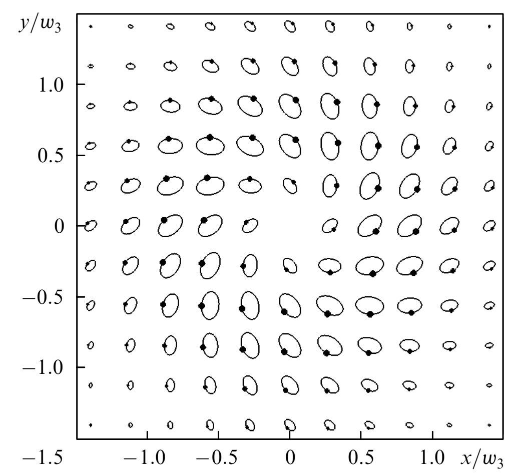

Figure 1. Intensity and polarisation distribution in a sum-frequency beam for the following parameters of incident radiation and nonlinear medium: $M_{01}=1$, $M_{02}=-1$, $b_1=b_3/3=b_4=b_5=-b_6/2=2b_7$, $\gamma_1=2\gamma_2/3=5\gamma_3/6=4\gamma_4/5=b_1$, $\chi=b_1$, $n_1=1.26$, $n_2=1.3$, $n_3=1.34$, $\omega_1=0.4\omega_3$, $w_1/w_2=2$.

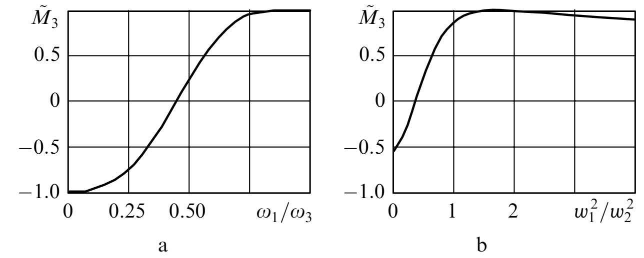

Figure 2. Dependences of $\tilde{M}_3(\omega_1/\omega_3)$ at $w_1/w_2 = 0.5$ (a) and $\tilde{M}_3(w_1^2/w_2^2)$ at $\omega_1/\omega_3 = 0.4$ (b); $M_{01} = 1$, $M_{02} = -1$, $b_1 = b_3/3 = b_4 = b_5 = -b_6/2 = 2b_7$, $\gamma_1 = 2\gamma_2/3 = 5\gamma_3/6 = 4\gamma_4/5 = b_1$, $\chi = b_1$, $n_1 = 1.26$, $n_2 = 1.3$, $n_3 = 1.34$.

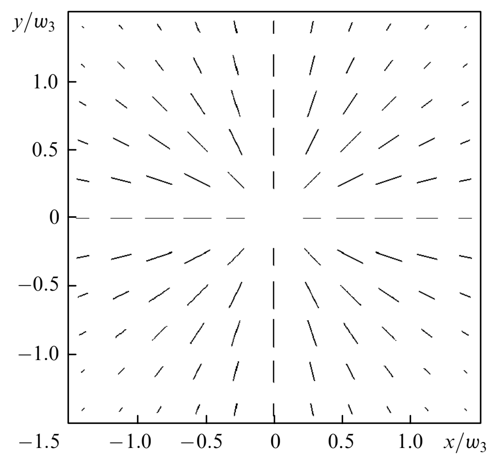

Figure 3. Intensity and polarisation distribution in a sum-frequency beam at $M_{01} = 1$, $M_{02} = -1$, $b_{1,2,3,\ldots,7} = 0$, $\gamma_1 = 2\gamma_2/3 = 5\gamma_3/6 = 4\gamma_4/5 = b_1$, $n_1 = 1.26$, $n_2 = 1.3$, $n_3 = 1.34$, $\omega_1 = 0.4\omega_3$, $w_1^2/w_2^2 = n_2\omega_2/(n_1\omega_1) = 1.548$.

$$+ \omega_3\,\chi(n_1 l_1 - n_2 l_2)/(n_1\omega_1 + n_2\omega_2 + n_3\omega_3)n_1 n_2 l_1 l_2)]$$

$$\times [((w_2^2\gamma_1 + w_1^2\gamma_2)/(w_1^2 + w_2^2) + (n_1\omega_1\gamma_1$$

$$+ n_2\omega_2\gamma_2)/n_3\omega_3)(n_1 b_4/l_1 + n_2 b_3/l_2) - n_3 b_1$$

$$\times (n_1 l_1 - n_2 l_2)(n_1\omega_1\gamma_3 - n_2\omega_2\gamma_4)/n_1 n_2 l_1 l_2\omega_3]^{-1}\}. \quad (21)$$

Figure 4a shows the intensity and polarisation distribution corresponding to this case in the plane of the reflected beam cross section at the sum frequency. One can clearly see four sectors obtained due to intersection of straight lines $\varphi = \varphi_{01}$ and $\varphi = \varphi_{02}$ at the point with the coordinates $(0, 0, z)$. Radiation within each of them is elliptically polarised and at the boundaries – linearly polarised. In this case, the rotation directions of the electric field vectors at the points lying in adjacent sectors are different (white and black ellipses).

The values of b_3, b_4, b_5 and b_6 can be found by realising the sum-frequency generation from the thin layer of chiral molecules deposited on the isotropic substrate ($\hat{\chi}^{(2)} \equiv 0$), the spatial dispersion of the quadratic nonlinearity of the material can be neglected. Consider first incident beams that are uniformly linearly polarised in the cross section plane with parallel polarisation planes ($\Psi_1 = \Psi_2 = 0$ and $M_{01} = M_{02} = 0$). In this case, $M_3(\varphi) \equiv 0$ and the distribution of the rotation angles of the principal axes of the polarisation ellipses at the sum frequency has a number of features (see Fig. 4b). In particular, two straight lines, $\varphi = \pi/2$ and $\varphi = \varphi^*$ exist in the beam cross section, where

$$\varphi^* = \arctan[(n_1\omega_2 w_2^2 b_6 - n_2\omega_1 w_1^2 b_5)$$

$$\times (n_1\omega_2 w_2^2 b_4 - n_2\omega_1 w_1^2 b_3)^{-1}]. \quad (22)$$

The electric field vector on these straight lines is collinear to them. n_1/n_2 and ω_1/ω_2 being invariable, a special choice of the ratio w_1/w_2 achieved by focusing the incident beams allows one to realise the situation, when $\varphi^* \approx \arctan(b_6/b_4)$ (at $w_1^2 \ll w_2^2$) or when $\varphi^* \approx -\arctan(b_5/b_3)$ (at $w_1^2 \gg w_2^2$). If now we change the polarisation of the incident beams into the circular one with the same handedness of the rotation direction of the electric field vector, other parameters being the same, the intensity $I_3(r, \varphi, z)$ will have the form

$$I_3(r, z_1) = 2|D(r, z)|^2(\omega_3 w_3^2)^2\{[n_2 b_3/(\omega_2 w_2^2)$$

$$+ n_1 b_4/(\omega_1 w_1^2)]^2 + [n_2 b_5/(\omega_2 w_2^2) - n_1 b_6/(\omega_1 w_1^2)]^2\}. \quad (23)$$

If $w_1^2 \ll w^2$, I_3 depends only on the components b_4 and b_6:

$$I_3(r, z) = 2|D(r, z_1)|^2 n_1^2(\omega_3^2/\omega_1^2)(b_4^2 + b_6^2). \quad (24)$$

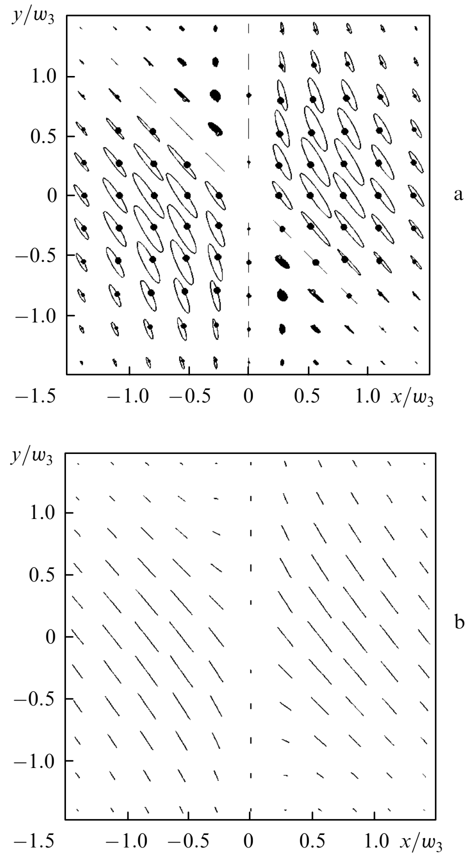

Figure 4. Intensity and polarisation distribution in a sum-frequency beam plotted for $w_1^2/w_2^2 = n_2\omega_2/n_1\omega_1 = 1.548$, $\chi = b_1$, $b_5 = b_1 = \gamma_1 = 2\gamma_2/3 = 5\gamma_3/6 = 4\gamma_4/5$ (a) and $b_5 = 2b_1$, $\gamma_{1,2,3,4} = 0$, $\chi = 0$, $w_1/w_2 = 0.1$ (b); $M_{01} = M_{02} = 0$, $\Psi_1 = \Psi_2 = 0$, $b_1 = b_3 = b_4/2 = -b_6/2 = 2b_7$, $n_1 = 1.26$, $n_2 = 1.3$, $n_3 = 1.34$, $\omega_1/\omega_3 = 0.4$, $\gamma_1 = 2\gamma_2/3 = 5\gamma_3/6 = 4\gamma_4/5 = b_1$.

Figure 4b was plotted for these parameters. Otherwise, when $w_1^2 \gg w_2^2$, the intensity I_3 has the form

$$I_3(r,z) = 2|D(r,z_1)|^2 n_1^2(\omega_3^2/\omega_1^2)(b_3^2 + b_5^2), \tag{25}$$

i.e. depends only on b_3 and b_5.

Thus, by using first linearly and then circularly polarised incident beams, i.e. experimentally determining the angle $\varphi^* \approx \arctan(b_6/b_4)$ [or $\varphi^* \approx -\arctan(b_5/b_3)$] and I_3 for different $\omega_{1,2}$, we can directly estimate the tensor components of the surface quadratic nonlinearity of the isotropic gyrotropic medium b_3, b_4, b_5 and b_6.

4. Conclusions

In this paper we have derived analytically expressions describing the intensity and polarisation distribution in the cross section of a sum-frequency beam reflected from the surface of an isotropic gyrotropic medium in the case of the normal incidence of elliptically polarised Gaussian fundamental beams. In calculations we have taken into account both the local and nonlocal contributions of the quadratic nonlinearity of the medium thickness and the nonlinear contribution of its surface in the generated signal. The corresponding selection of polarisations of fundamental waves makes it possible to distinguish the contribution of the medium surface from the contribution of its volume. The peculiarities of the polarisation distribution appearing in this case in the cross section plane of the signal beam allow one to draw conclusion about the character of the nonlinear response of the medium and in a number of cases extract quantitative information about tensor components characterising the quadratic nonlinearity of the surface.

We should keep in mind that the polarisation distribution in the sum-frequency beam can rather strongly vary in a small range. This, on the one hand, can complicate interpretation of experimental results (the discussion of the possibility to observe transverse intensity and polarisation distributions similar to that described in this paper is presented in [5, 6]) and, on the other hand, makes it possible to speak about the possible vistas of the effect considered from the point of view of formation of nonuniformly polarised light beams.

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