NONLINEAR OPTICAL PHENOMENA

PACS numbers: 42.65.Tg; 42.65.Dr.; 78.20.Fm
DOI: 10.1070/QE2005v035n11ABEH008993

Continuous Stokes self-scattering of an optical pulse in a uniaxial crystal in the case of Zakharov – Benney resonance

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***Abstract*. A new mechanism of the continuous frequency down-conversion of a pulse propagating in a uniaxial crystal under conditions of the Zakharov – Benney resonance between its ordinary and extraordinary components is proposed. Unlike stimulated Raman self-scattering, here the red frequency shift of the ordinary component saturates, achieving its maximum value at some propagation length, which is proportional to the input pulse intensity. The ordinary component generates an ultimately short single pulse of the extraordinary wave propagating in the soliton regime. It is shown that this effect can be observed only in a crystal with the positive birefringence.**

Keywords: *Stokes self-scattering, Zakharov – Benney resonance, uniaxial crystal, soliton.*

1. Introduction

The discovery of different mechanisms of nonlinear frequency conversion of coherent optical pulses stimulated the development of dynamically tunable lasers [1]. One of the most efficient mechanisms of continuous frequency conversion is stimulated Raman scattering (SRS) [2 – 5]. This effect is observed upon irradiation of a Raman-active medium by femtosecond laser pulses. Because of the short duration τ_p of these pulses, their spectrum of width $\delta\omega \sim 1/\tau_p$ contains the carrier frequency ω and the Stokes component $\omega - \omega_v$, where ω_v is the frequency of an optical vibrational mode of Raman-active molecules. As a result, during the propagation of femtosecond pulses, the oscillations of optical vibrational modes of the medium are coherently enhanced, which is accompanied by a continuous change of the spectrum of optical signals. When the role of dispersion is substantial, the soliton SRS regime is realised at which the spectrum of the pulse shifts to the red as a whole. This shift increases with increasing the pulse intensity [5] and can considerably exceed ω_v. Therefore, SRS is the result of nonlinear interaction between the field of a short light pulse and vibrational modes of molecules of the medium. It is known that the action of this field on modes is determined by the square of the field strength.

The field of a light pulse in uniaxial crystals has two components: ordinary E_o and extraordinary E_e, which can efficiently interact with each other under certain conditions due to the quadratic nonlinearity. For picosecond pulses considered in this paper, the quadratic nonlinearity in uniaxial crystals is mainly determined by the nonresonance electronic transitions [6]. In this case, the spectrum of the pulse lies in the transparency region of the crystal.

The quadratic nonlinearity was successfully used for continuous parametric frequency conversion under phase-matching conditions for three-wave mixing [7 – 10], which are determined by the laws of conservation of energy and momentum,

$$\omega_1 = \omega_2 + \Omega, \quad \boldsymbol{k}^{(1)} = \boldsymbol{k}^{(2)} + \boldsymbol{k}^{(3)}, \tag{1}$$

where ω_1 is the frequency of the optical (pump) wave incident on a crystal, which decomposes inside the crystal into two waves with frequencies ω_2 and Ω (the signal and idler components, respectively); and $\boldsymbol{k}^{(1)}$, $\boldsymbol{k}^{(2)}$, $\boldsymbol{k}^{(3)}$ are the wave vectors corresponding to these frequencies.

By varying the angle between $\boldsymbol{k}^{(1)}$ and the optical axis, we can continuously vary the frequencies ω_2 and Ω for which phase-matching conditions (1) are valid. For short pulses incident on a crystal, the frequencies ω_1 and ω_2 can be initially contained in their spectrum if these frequencies are sufficiently close to each other so that $\omega_1 - \omega_2 \sim 1/\tau_p \ll \omega_1, \omega_2$. The Cherenkov mechanism of generation of a single-cycle IR pulse (video pulse) proposed in [11] and realised experimentally in [12] is based on this property. In these papers, quasi-monochromatic pulses generating a video pulse were powerful enough, so that their field was assumed specified. In the case of self-consistent approach to the solution of this problem, it is necessary to take into account the reverse action of the video pulse on the pulse generating it. Therefore, we can speak about the self-scattering of a quasi-monochromatic pulse in a medium with a quadratic optical nonlinearity.

Here, an analogy with the above-mentioned interaction between vibrational molecular modes and an optical pulse exists. In this case, the role of a generated video pulse is similar to that of the optical vibrational molecular mode in SRS. The important difference is that the group velocity of vibrational modes is virtually zero (i.e., they cannot propagate themselves), whereas the group velocity of the extraordinary wave only slightly differs from that of the ordinary wave. For this reason, the conditions for efficient energy transfer from the ordinary component to the

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Received 18 April 2005; revision received 13 July 2005
Kvantovaya Elektronika **35** (11) 1019 – 1026 (2005)
Translated by M.N. Sapozhnikov

extraordinary one should differ from the conditions for SRS realisation. At the same time, the similarity between these two types of nonlinear interaction pointed out above suggests that the frequency (or spectrum) of a pulse propagating in a nonlinear uniaxial crystal can be tuned in principle. In this case, the nonlinearity has the electronic nature rather than Raman. In turn, the pulse is split into two different interacting components due to the medium anisotropy.

This paper is devoted to the study of self-scattering of quasi-monochromatic pulses by video pulses generated by them in quadratically nonlinear optically homogeneous crystals.

2. Nonlinear wave equations and soliton regime of Stokes self-scattering

We assume that an ordinary wave is incident on a crystal. Before proceeding to the derivation of nonlinear wave equations, we consider relations (1) in more detail. By denoting the angle between $\boldsymbol{k}^{(1)}$ and $\boldsymbol{k}^{(2)}$ by θ, we obtain from the second relation

$$k^{(3)2} = (k^{(1)} - k^{(2)})^2 + 4k^{(1)}k^{(2)}\sin^2\frac{\theta}{2}. \tag{2}$$

As pointed out in Introduction, we assume that the frequencies ω_1 and ω_2 are close to each other and are contained in the spectrum of the initial pulse. Then, we can write

$$k^{(1)} - k^{(2)} \approx \left(\frac{\partial k}{\partial\omega}\right)_{\omega=\omega_1}(\omega_1 - \omega_2) = \frac{\Omega}{v_g},$$

where v_g is the group velocity of the pulse. Taking also into account that $k^{(1)} = n_1\omega_1/c$, $k^{(3)} = n_3\Omega/c$ [c is the speed of light in vacuum, $n_1 = n_1(\omega_1)$ and $n_3 = n_3(\Omega)$ are the refractive indices at the corresponding frequencies], we rewrite (2) in the form

$$\left(\frac{n_3^2}{c^2} - \frac{1}{v_g^2}\right)\Omega^2 = 4\frac{n_1\omega_1}{c}\left(\frac{n_1\omega_1}{c} - \frac{\Omega}{v_g}\right)\sin^2\frac{\theta}{2}. \tag{3}$$

where $\Omega \ll \omega_1$, as mentioned above.

Expression (3) can be considered as the equation describing the dependence of Ω on the angle θ and carrier frequency ω_1 of the incident pulse. Note that in the collinear regime ($\theta = 0$) and under the condition

$$v_g = \frac{c}{n_3} \tag{4}$$

Eqn (3) turns to the identity and Ω can take any value, i.e., becomes a free parameter. Therefore, under these conditions, the value of Ω cannot be determined from the conservation laws in elementary scattering events. Obviously, Ω is the red shift of the frequency ω_1 of the initial pulse propagating in a nonlinear medium.

Condition (4) corresponds to the limiting case of three-wave interaction, when $\omega_1, \omega_2 \gg \Omega$ and $k^{(1)}, k^{(2)} \gg k^{(3)}$. Indeed, under these conditions, we find from (3)

$$\frac{\Omega}{k^{(3)}} = \frac{c}{n_3} = \frac{\omega_1 - \omega_2}{k^{(1)} - k^{(2)}} \approx \left(\frac{\partial\omega}{\partial k}\right)_{\omega=\omega_1} = v_g,$$

i.e., the phase velocity of the low-frequency wave is equal to the group velocity of the high-frequency wave. In the theory of nonlinear waves, this condition is called the Zakharov–Benney resonance (ZBR) [13, 14].

We assume that a laser beam, whose polarisation plane coincides with that of the ordinary wave, is incident on a crystal along the z axis, which forms an angle φ with the optical axis c (Fig. 1). We also assume that the spectrum of the pulse lies in the transparency region of the crystal, where dispersion can be assumed relatively small [15]. For this reason, we will consider dispersion only in the linear part of the medium response. The quadratic nonlinearity will be taken into account by means of the second-order nonlinear susceptibility tensors. The wave equations can be written in the form

$$\Delta E_{e,o} - \frac{1}{c^2}\frac{\partial^2 E_{e,o}}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}P_{e,o}, \tag{5}$$

where Δ is the Laplace operator.

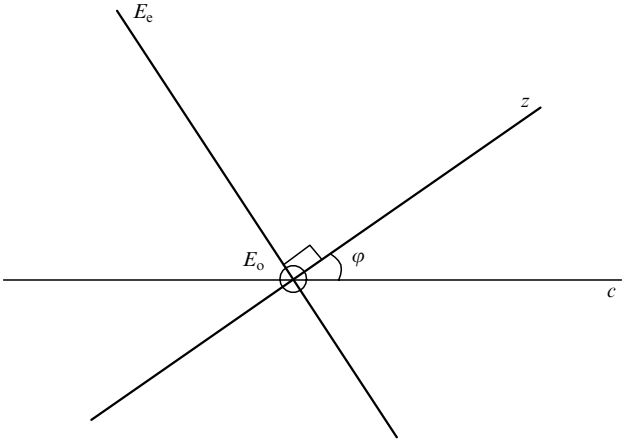

Figure 1. Propagation geometry of an optical pulse in a uniaxial crystal: c is the optical axis; z is the pulse propagation direction.

We write polarisations P_o and P_e in the form

$$P_o = \int_0^\infty \tilde{\chi}_o(\tau)E_o(t-\tau)d\tau + 2\chi_{eo}E_eE_o, \tag{6}$$

$$P_e = \int_0^\infty \tilde{\chi}_e(\tau)E_e(t-\tau)d\tau + \chi_{eo}E_o^2 + \chi_{ee}E_e^2, \tag{7}$$

where $\tilde{\chi}_o(\tau) = \chi_{xx}^{(1)}(\tau)$, and $\tilde{\chi}_e(\tau) = \chi_{yy}^{(1)}(\tau)$ are the ordinary and extraordinary components of the linear electronic susceptibility tensor taking into account the time delay of the response (dispersion); and $\chi_{eo} = \chi_{xxy}^{(2)} = \chi_{xyx}^{(2)} = \chi_{yxx}^{(2)}$, and $\chi_{ee} = \chi_{yyy}^{(2)}$ are the nonzero components of the inertialless quadratic susceptibility.

Note that the symmetry properties of a uniaxial medium, namely, the invariance with respect to spatial reflection normal to the optical axis are taken into account in (6) and (7). Because the components E_o and P_o are perpendicular to this axis and are polar vectors, $P_o \to -P_o$, and $E_o \to -E_o$ upon these reflections. In the case of spatial inversion in any other directions, this invariance is violated and, hence, expressions (6) and (7) are not invariant with respect to the $P_e \to -P_e$, $E_e \to -E_e$ transformation (see Fig. 1).

Crystals of the medium symmetry class are optically uniaxial. This symmetry includes three syngonies: trigonal, tetragonal, and hexagonal. Crystals of these syngonies have the same structure of the linear susceptibility tensor reduced to the principal axes [16]. The primitive unit cells have no cylindrical symmetry and possess only symmetry for rotations through angles 120°, 90° and 60°. However, the optical properties of these crystals can be described by treating them as axially symmetric media. In this case, the number of nonzero components of nonlinear susceptibility is insensitive to the syngony type because the wavelength in the optical range considerably exceeds interatomic distances.

By substituting (6) and (7) into (5), we arrive at the system of nonlinear wave equations

$$\Delta E_{\rm o} - \frac{1}{c^2}\frac{\partial^2 E_{\rm o}}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\left[\int_0^\infty \tilde{\chi}_{\rm o}(\tau)E_{\rm o}(\boldsymbol{r}, t-\tau)\mathrm{d}\tau + 2\chi_{\rm eo}E_{\rm e}E_{\rm o}\right], \tag{8}$$

$$\Delta E_{\rm e} - \frac{1}{c^2}\frac{\partial^2 E_{\rm e}}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\left[\int_0^\infty \tilde{\chi}_{\rm e}(\tau)E_{\rm e}(\boldsymbol{r}, t-\tau)\mathrm{d}\tau + \chi_{\rm eo}E_{\rm o}^2 + \chi_{\rm ee}E_{\rm e}^2\right]. \tag{9}$$

Because the component $E_{\rm o}$ has the carrier frequency ω, it can be written in the form

$$E_{\rm o}(\boldsymbol{r}, t) = \psi(\boldsymbol{r}, t)\exp[\mathrm{i}(\omega t - kz)] + \text{c.c.}, \tag{10}$$

where $\psi(\boldsymbol{r}, t)$ is the slowly varying envelope and k is the wave number.

As pointes out above, the ordinary component is incident on the crystal. In this case, as follows from (6) and (7), the extraordinary component $E_{\rm e}$ can be centred either at the frequency 2ω or zero frequency. The first case corresponds to the known effect of the second harmonic generation, and the second one, under certain conditions, to the generation of a video pulse [7, 17]. It is in the second case that some similarity to the SRS theory can be found [5], where normal optical vibrational modes, unlike the light field, have no carrier frequency. The difference is that the group velocity of optical vibrational molecular modes is virtually zero, whereas for the component $E_{\rm e}$ it is close to the velocity of the ordinary wave $E_{\rm o}$.

According to [18], due to a slow variation of the envelope ψ at the time scale τ, we perform the expansion into a Taylor series in τ:

$$\int_0^\infty \tilde{\chi}_{\rm o}(\tau)E_{\rm o}(\boldsymbol{r}, t-\tau)\mathrm{d}\tau = \left[\chi_{\rm o}(\omega)\psi - \mathrm{i}\left(\frac{\partial\chi_{\rm o}}{\partial\omega}\right)\frac{\partial\psi}{\partial t} - \frac{1}{2}\left(\frac{\partial^2\chi_{\rm o}}{\partial\omega^2}\right)\frac{\partial^2\psi}{\partial t^2}\right]\exp[\mathrm{i}(\omega t - kz)] + \text{c.c.}, \tag{11}$$

where the frequency susceptibility is

$$\chi_{\rm o}(\omega) = \int_0^\infty \tilde{\chi}_{\rm o}(\tau)\mathrm{e}^{-\mathrm{i}\omega\tau}\mathrm{d}\tau.$$

Because of a relatively weak dispersion, the integrand in the right-hand side of (9) can be also expanded in τ:

$$\int_0^\infty \tilde{\chi}_{\rm e}(\tau)E_{\rm e}(\boldsymbol{r}, t-\tau)\mathrm{d}\tau = \chi_{\rm e}(0)E_{\rm e}(\boldsymbol{r}, t) - \chi'_{\rm e}(0)\frac{\partial E_{\rm e}(\boldsymbol{r}, t)}{\partial t} - \frac{\chi''_{\rm e}(0)}{2}\frac{\partial^2 E_{\rm e}(\boldsymbol{r}, t)}{\partial t^2}, \tag{12}$$

where

$$\chi_{\rm e}(0) = \int_0^\infty \chi_{\rm e}(\tau)\mathrm{d}\tau$$

is the inertialless part of the susceptibility;

$$\chi'_{\rm e}(0) = \int_0^\infty \tau\chi_{\rm e}(\tau)\mathrm{d}\tau; \qquad \chi''_{\rm e}(0) = -2\int_0^\infty \tau^2\chi_{\rm e}(\tau)\mathrm{d}\tau.$$

The term $\sim \partial E_{\rm e}/\partial t$ in (12) describes the decay of the polarisation response of a medium caused by irreversible relaxation with the characteristic time T_2, which is determined by the coefficient $\chi'_{\rm e}(0)$. For a picosecond pulse and a broad class of crystals, the condition $\tau_{\rm p} \ll T_2$ is fulfilled with good margin, so that the term $\sim \partial E_{\rm e}/\partial t$ in (12) can be neglected. To confirm this, we will use the Sellmeyer formula

$$\chi_{\rm e} = \chi_{\rm e}(0)\frac{\omega_{\rm e}^2}{\omega_{\rm e}^2 - \mathrm{i}\gamma\tilde{\omega} - \tilde{\omega}^2},$$

where $\omega_{\rm e}$ is the characteristic frequency of optical electronic transitions excited by the extraordinary wave; $\gamma = 1/T_2$; and $\tilde{\omega}$ is the characteristic frequency in the $E_{\rm e}$ spectrum.

For the transparency region ($\tilde{\omega}^2 \ll \omega_{\rm e}^2$), the expansion

$$\chi_{\rm e} = \chi_{\rm e}(0)\left(1 + \mathrm{i}\gamma\frac{\tilde{\omega}}{\omega_{\rm e}^2} + \frac{\tilde{\omega}^2}{\omega_{\rm e}^2}\right)$$

is valid.

By making substitution $\tilde{\omega} \to \mathrm{i}\partial/\partial t$ and using the relation $P_{\rm e} = \chi_{\rm e}E_{\rm e}$, we obtain (12).

Let us emphasise once more that expansions (11) and (12) have different physical substantiations because ψ in (11) is the envelope of the ordinary wave, whereas $E_{\rm e}$ in (12) is the field itself of the extraordinary component.

By substituting (11) and (12) into (8) and (9) and neglecting the relatively rapidly oscillating terms, we obtain

$$\mathrm{i}\left(\frac{\partial\psi}{\partial z} + \frac{1}{v_{\rm g}}\frac{\partial\psi}{\partial t}\right) + \frac{k_2}{2}\frac{\partial^2\psi}{\partial t^2} = \beta_{\rm eo}E_{\rm e}\psi + \frac{c}{2n_{\rm o}\omega}\Delta_\perp\psi, \tag{13}$$

$$\frac{\partial^2 E_{\rm e}}{\partial z^2} - \frac{n^2}{c^2}\frac{\partial^2 E_{\rm e}}{\partial t^2} = \frac{\partial^2}{\partial t^2}(\alpha_{\rm eo}|\psi|^2 + \alpha_{\rm ee}E_{\rm e}^2) - \delta_{\rm e}\frac{\partial^4 E_{\rm e}}{\partial t^4} - \Delta_\perp E_{\rm e}, \tag{14}$$

where $v_{\rm g} = c/[n_{\rm o} + \omega(\partial n_{\rm o}/\partial\omega)]$ is the group velocity of the ordinary wave; $n_{\rm o} = n_{\rm o}(\omega) = [1 + 4\pi\chi_{\rm o}(\omega)]^{1/2}$ and $n = [1 + 4\pi\chi_{\rm e}(0)]^{1/2}$ are the refractive indices for the ordinary and extraordinary waves, respectively; $k_2 = \partial(1/v_{\rm g})/\partial\omega$ is the group-velocity dispersion (GVD) parameter of the ordinary wave; $\beta_{\rm eo} = 2\pi\omega\chi_{\rm eo}/c$, $\alpha_{\rm eo} = 8\pi\chi_{\rm eo}/c^2$, and $\alpha_{\rm ee} = 4\pi\chi_{\rm ee}/c^2$ are the nonlinearity coefficients; $\delta_{\rm e} = 2\pi\chi''_{\rm e}(0)/c^2$ is the

dispersion parameter of the extraordinary wave; and $\Delta_\perp$ is the transverse Laplacian.

One can see from system (13), (14) that the envelope ψ of the ordinary wave plays a dominant role: If a pulse polarised in the ordinary-wave plane is incident on a medium, a video pulse of the extraordinary wave is generated in the medium due to nonlinearity [the second term in the parentheses in the right-hand part of (14)]; at the same time, it follows from (13) that the extraordinary wave cannot generate the ordinary wave in the absence of the latter at the input to the medium. This asymmetry is inherent in uniaxial crystals and is caused by the above-mentioned invariance of equations with respect to the $E_o \to -E_o$ operation and its violation for $E_e \to -E_e$. For this reason, we can neglect the intrinsic dispersion and nonlinearity of the extraordinary wave in the system of equations (13), (14) as the higher smallness order effects.

The ordinary wave makes the main contribution to dispersion [the last term in the left-hand side of (13)]. In the right-hand side of (14), we retain only the nonlinearity $\partial^2(\alpha_{eo}|\psi|^2)/\partial t^2$ produced by the ordinary wave. In addition, it is convenient for a further analysis to pass in (14) to the quasi-uniaxial propagation approximation [14]. This is possible because nonlinearity in this case is represented by a power series in the field strength. Thus, we have in the right-hand side of (14) the terms of the higher smallness order, which allows us to neglect a wave reflected from the inhomogeneities of the medium produced due to nonlinearity. Summarising the above said, we transform (13) and (14) to the system

$$\mathrm{i}\frac{\partial\psi}{\partial z}+\frac{k_2}{2}\frac{\partial^2\psi}{\partial\tau^2}=\beta_{eo}E_e\psi+\frac{c}{2n_o\omega}\Delta_\perp\psi,$$
$$\frac{\partial E_e}{\partial z}=-\alpha\frac{\partial}{\partial\tau}(|\psi|^2)+\frac{c}{2n}\Delta_\perp\int_{-\infty}^{\tau}E_e\mathrm{d}\tau', \tag{15}$$

where $\alpha = c\alpha_{eo}/(2n)$, $\tau = t - nz/c = t - z/v_g$. Here, we took into account condition (4).

In the absence of transverse perturbations, system (15) transforms to the integrable Yadjima–Oikawa system [19], which has the single-soliton solution of the type

$$\psi=\psi_m\exp[-\mathrm{i}(\Omega t-qz)]\mathrm{sech}\left(\frac{t-z/v}{\tau_p}\right),$$
$$E_e=E_{em}\mathrm{sech}^2\left(\frac{t-z/v}{\tau_p}\right), \tag{16}$$

where

$$\psi_m=\frac{|k_2|}{\tau_p}\left(\frac{\Omega}{\alpha\beta_{eo}}\right)^{1/2}; \tag{17}$$

$E_{em} = -k_2/(\beta_{eo}\tau_p^2)$; and the propagation velocity v is described by the expression

$$\frac{1}{v}=\frac{n_e}{c}-k_2\Omega.$$

Solution (16) depends on two free parameters: the duration τ_p and nonlinear frequency shift Ω of the ordinary component of a soliton. It follows from expression (17) that $\Omega \geqslant 0$. This and (16) and (10) suggest that the ordinary component shifts to the red ($\omega \to \omega - \Omega$). This shift can be treated as a continuous red shift of the ordinary component upon energy transfer from this component to the extraordinary wave representing a unipolar video pulse [see the second expression in (16)]. Finally, this process is stabilised, and a soliton of type (16) with the constant frequency shift Ω is formed. We can say that during the formation of an ordinary–extraordinary soliton, the Stokes frequency shift saturates. From the physical point of view, this saturation is explained by the fact that condition (4) is violated during the frequency shift, resulting in a decrease in the efficiency of energy transfer from the ordinary to extraordinary wave.

The value of Ω depends on the input parameters of an optical pulse and can be determined by solving the corresponding inverse scattering problem. Another method for determining the frequency shift involves a direct numerical integration of (13) and (14) or (15). Note here that, as follows from expression (17) $\Omega \sim \psi_m^2 \sim I_o$ where I_o is the ordinary component intensity. This means that the value of the red shift is proportional to the intensity of the input pulse polarised in the ordinary wave plane. As pointed out in Section 2 for $\theta = 0$ and condition (4), the value of Ω cannot be determined from the laws of conservation of energy and momentum in elementary photon–photon scattering events. Because $I_o \sim n_{ph}$, where n_{ph} is the concentration of photons forming the ordinary component of the pulse, we can say that Ω is determined under given conditions by collective scattering events rather than elementary events. A similar situation takes place upon SRS [2–5, 20]. By using expression (17), expressions for α, β_{eo} and the estimate $k_2 \sim (c\omega)^{-1}$, we obtain

$$\frac{\Omega}{\omega}\sim\frac{32\pi^3}{c}I_o\chi_{eo}^2(\omega\tau_p)^2. \tag{18}$$

By taking $\tau_p \sim 1$ ps, $I_o \sim 10^{11}$ W cm^{-2}, $\omega \sim 10^{15}$ s^{-1}, and $\chi_{eo} \sim 10^{-9}$ esu [21], we find $\Omega/\omega \sim 0.1$. Thus, in the case of ZBR in a uniaxial crystal, quite efficient frequency conversion of the input pulse to the red can be expected.

Let us estimate under these conditions the intensity of the video pulse of the ordinary wave:

$$I_e \cong \frac{E_{em}^2}{\psi_m^2}I_o \sim \frac{I_o}{\omega\Omega\tau_p^2}.$$

For the parameters presented above, we have $I_e \sim 10^{-5}I_o \sim 10^6$ W cm^{-2}, i.e., the intensity of the extraordinary component is considerably lower than that of the ordinary component. This, by the way, well justifies the neglect of the intrinsic nonlinearity of the extraordinary wave and its dispersion in (13) and (14).

Let us determine the region of values of carrier frequencies of the pulses incident on a crystal at which condition (4) can be fulfilled. Note that (4) can be rewritten in the form

$$n(0,\varphi)=n_o(\omega)+\omega\frac{\partial n_o}{\partial\omega}. \tag{19}$$

At the same time, the dependence of n on φ is determined by the expression [22]

$$\frac{1}{n^2}=\frac{\cos^2\varphi}{n_o^2}+\frac{\sin^2\varphi}{n_e^2}, \tag{20}$$

where n_e is the extraordinary refractive index equal to n for $\varphi = 90^\circ$. Equations (19) and (20) determine in the general case the dependence of the carrier frequency of the input pulse on the angle φ at which its Stokes self-scattering is observed.

Note first that $\partial n_o/\partial\omega > 0$ in the transparency region of the medium. Therefore, the Stokes self-scattering, as one can see from (19), can occur only in crystals with the positive birefringence ($n_e > n_o$). In addition, in the transparency region the expansion

$$n_o^2(\omega) - 1 = A\left[1 + a\frac{\omega^2}{\omega_0^2} + \ldots - \left(\frac{\omega_{pi}}{\omega_{pe}}\right)^2 a'\frac{\omega_0^2}{\omega^2} - \ldots\right]$$

is valid [15], where A is a constant related to the inertialless part $\tilde{n}_o$ of the refraction index by the expression $A = \tilde{n}_o^2 - 1$; ω_{pe} and ω_{pi} are the electron and ion plasma frequencies, respectively; ω_0 is the characteristic frequency of optical electronic transitions; and $a, a' \ldots$ are empirical constants of the order of unity. The group of terms in the right-hand side with the '+' sign describes the electronic response of the crystal, while the group of terms with the '–' sign describes the ion response.

Because the above expression is a power series, we have $a\omega^2/\omega_0^2, (\omega_{pi}/\omega_{pe})^2 a'\omega_0^2/\omega^2, \ldots \ll 1$. Then, this expression can be approximately rewritten in the form

$$n_o(\omega) = \tilde{n}_o + \frac{\tilde{n}_o^2 - 1}{2\tilde{n}_o}\left[3a\frac{\omega^2}{\omega_0^2} - \left(\frac{\omega_{pi}}{\omega_{pe}}\right)^2 a'\frac{\omega_0^2}{\omega^2}\right]. \qquad (21)$$

By substituting (21) into (19), we rewrite the ZBR condition in the form

$$n(0) - \tilde{n}_o = \frac{\tilde{n}_o^2 - 1}{2\tilde{n}_o}\left[9a\frac{\omega^2}{\omega_0^2} + \left(\frac{\omega_{pi}}{\omega_{pe}}\right)^2 a'\frac{\omega_0^2}{\omega^2}\right]. \qquad (22)$$

By solving this equation for ω, we find the carrier frequencies of pulses converted to the red region in the crystal:

$$\omega_\pm^2 = \frac{\tilde{n}_o^2\omega_0^2}{9a(\tilde{n}_o^2 - 1)}\left\{n - \tilde{n}_o \pm \left[(n - \tilde{n}_o)^2 - 9aa'\right.\right.$$
$$\left.\left.\times\left(\frac{\omega_{pi}}{\omega_{pe}}\right)^2\left(\frac{\tilde{n}_o^2 - 1}{\tilde{n}_o}\right)^2\right]^{1/2}\right\}. \qquad (23)$$

From the condition that the expression in brackets should be positive, we obtain the restriction

$$n - \tilde{n}_o > 3\frac{\omega_{pi}}{\omega_{pe}}\frac{\tilde{n}_o^2 - 1}{\tilde{n}_o}(aa')^{1/2}. \qquad (24)$$

By using (20), we rewrite (24) in the form

$$\sin^2\varphi > \sin^2\varphi_{min} = 3\frac{\omega_{pi}}{\omega_{pe}}\frac{\tilde{n}_o^2 - 1}{\tilde{n}_o(n_e - \tilde{n}_o)}(aa')^{1/2}. \qquad (25)$$

Therefore, we have the restriction from below on the angle between the optical axis and the pulse propagation direction. For this reason, the discarding of cubic nonlinearities in (6) and (7) is justified. These nonlinearities can dominate at small values of φ (for $\varphi = 0$, the quadratic susceptibilities are strictly zero due the axial symmetry).

In addition, it follows from (23) that the value of $\omega_\pm$ depends on φ. Therefore, the ZPR condition can be obtained by varying φ and the carrier frequency of the input pulse of the ordinary wave. Because the dependence $n(\varphi)$ in crystals with the positive birefringence monotonically increases, the frequency ω_+ increases with increasing φ, achieving the maximum value for $\varphi = 90^\circ$, which is determined from (18) upon the substitution $n \to n_e$. At the same time, one can easily see that ω_- decreases with increasing φ (Fig. 2).

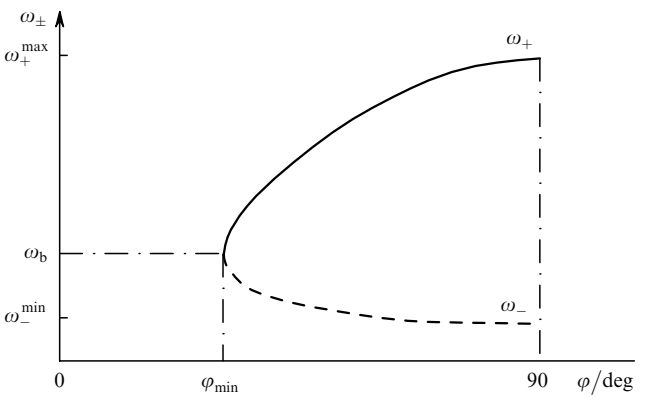

Figure 2. Relation between the carrier frequency $\omega_\pm$ of the input pulse and the angle φ between its direction and the optical axis upon the efficient Stokes conversion in the ZBR regime. The branch ω_+ (solid curve) corresponds to the positive GVD, the branch ω_- (dashed curve) – to the negative GVD. Nonlinearity has a defocusing nature in the spectral range $\omega_b < \omega < \omega_+^{max}$ and a focusing nature in the range $\omega_-^{min} < \omega < \omega_b$, where the boundary frequency is $\omega_b = \omega_0\tilde{n}_o/3[(n - \tilde{n}_o)/a(\tilde{n}_o^2 - 1)]^{1/2}$.

Assuming that $a, a' \sim 1, \omega_{pi}/\omega_{pe} \sim 10^{-2}, (\omega/\omega_0)^2 \sim 10^{-2}$ and $\tilde{n}_0^2 \cong 2$, we obtain $n_e - n_o > 10^{-2} - 10^{-3}$, which is realised for a broad class of uniaxial crystals [23].

We will call the carrier frequencies ω_+ and ω_- determined by (23) the high and low frequencies, respectively. Estimates show that $\omega_+ \sim 10^{15}$ s^{-1}, $\omega_- \sim 10^{14}$ s^{-1}, i.e., the high frequency belongs to the visible range, while the low frequency lies in the near-IR range. The GVD parameter k_2 has the form

$$k_2 = \frac{\partial}{\partial\omega}\left(\frac{1}{v_g}\right) = 3a\omega\frac{\tilde{n}_o^2 - 1}{\tilde{n}_o}\left[1 - \frac{a'}{a}\left(\frac{\omega_{pi}}{\omega_{pe}}\right)^2\frac{\omega_0^4}{\omega^4}\right]. \qquad (26)$$

It follows from (26) and (23) that the GVD in the medium is positive ($k_2 > 0$) for ω_+ and negative ($k_2 < 0$) for ω_-. This circumstance, as shown below, is important in the consideration of the influence of transverse perturbations.,

3. Consideration of transverse perturbations

From the point of view of possible experiments, the stability of a soliton (16) with respect to transverse perturbations is important. Let us take these perturbations into account by the Whitham method of the 'averaged Lagrangian' [24].

To system (15), the density of the Lagrangian

$$L = \frac{i}{2}\left(\psi\frac{\partial\psi^*}{\partial z} - \psi^*\frac{\partial\psi}{\partial z}\right) + \frac{k_2}{2}\left|\frac{\partial\psi}{\partial\tau}\right|^2 - \beta_{eo}|\psi|^2\frac{\partial u}{\partial\tau}$$

$$-\frac{c}{2n_o\omega}|\nabla_\perp\psi|^2+\frac{\beta_{eo}}{2\alpha}\left[\frac{\partial u}{\partial z}\frac{\partial u}{\partial\tau}-\frac{c}{2n}(\nabla_\perp u)^2\right] \tag{27}$$

corresponds, where the variable u is related to the field strength of the extraordinary wave by the expression $E_e=\partial u/\partial\tau$.

By using one-dimensional solitons (16), we will use the expressions

$$\psi=|k_2|\left(\frac{\Omega}{\alpha\beta_{eo}}\right)^2\rho\exp\left[-i\Omega\left(t-\frac{nz}{c}\right)-\frac{n_o\omega}{c}\Phi\right]\operatorname{sech}\left[\rho\left(t-\frac{z}{v}\right)\right], \tag{28}$$

$$u=-\frac{k_2}{\beta_{eo}}\rho\tanh\left[\rho\left(t-\frac{z}{v}\right)\right]$$

as trial solutions that take transverse perturbations into account. Here, new dynamic parameters ρ and Φ, which correspond to the inverse duration of the soliton and the eikonal of its ordinary component, respectively, are the functions of coordinates that should be found, and the parameter Ω is still assumed constant in the formed soliton.

By substituting (28) into (27) and integrating with respect to the fast variable τ, as was done in [25] for other nonlinear equations, we obtain the 'averaged Lagrangian' of the form

$$\Lambda\equiv\frac{c\alpha\beta_{eo}}{2n_o\omega\Omega k_2^2}\int_{-\infty}^{+\infty}L\,d\tau=-\rho\frac{\partial\Phi}{\partial z}-\frac{1}{2}\rho(\nabla_\perp\Phi)^2+\frac{ck_2}{2n_o\omega}(\Omega^2\rho-\rho^3/3)-b\frac{(\nabla_\perp\rho)^2}{4\rho}, \tag{29}$$

where

$$b=\frac{c}{3n_o\omega}\left[\left(\frac{\pi^2}{6}+2\right)\frac{c}{n_o\omega}+\frac{c}{2\Omega n}\right]\approx\frac{c^2}{6n_o n\omega\Omega}. \tag{30}$$

By writing the Euler–Lagrange equations for ρ and Φ with the use of (29), we obtain the system of the 'hydrodynamic' type

$$\frac{\partial\rho}{\partial z}+\nabla_\perp(\rho\nabla_\perp\Phi)=0,$$
$$\frac{\partial\Phi}{\partial z}+\frac{(\nabla_\perp\Phi)^2}{2}+\frac{ck_2}{2n_o\omega}(\rho^2-\Omega^2)=b\frac{\Delta_\perp\sqrt{\rho}}{\sqrt{\rho}}. \tag{31}$$

In the one-dimensional case, system (31) has the solutions $\rho=1/\tau_p=\text{const}$, $\Phi=\Phi_0(z)=(ck_2/2n_o\omega)(\Omega^2-1/\tau_p^2)\times z$, which exactly correspond to the soliton solution (16). This is an important argument in favour of the 'averaged Lagrangian' method.

Transverse perturbations are taken into account in the left- and right-hand sides of the second equation in (31). It follows from (30) that the coefficient b tends to zero for $\omega\sim1/\lambda\to\infty$, where λ is the wavelength of the ordinary component of the soliton. The tendency of the wavelength to zero corresponds to the eikonal approximation (geometrical optics approximation). Therefore, the term $b\Delta_\perp\sqrt{\rho}/\sqrt{\rho}$ in the right-hand side of the second equation in (31) describes the wave properties in the transverse dynamics of the soliton (diffraction effects), while the term $(\nabla_\perp\Phi)^2/2$ in the left-hand side describes the transverse dynamics in the eikonal approximation, i.e., nonlinear refraction.

Consider first the stability of a soliton with respect to small-scale perturbations by representing ρ and Φ in the form $\rho=1/\tau_p+\rho_1$ and $\Phi=\Phi_0+\Phi_1$, where $\rho_1\ll1/\tau_p$, $\Phi_1\ll\Phi_0$. By linearising (31) with respect to ρ_1, Φ_1 and assuming that $\rho_1,\Phi_1\sim\exp[i(q_\parallel z+\boldsymbol{q}_\perp\boldsymbol{r}_\perp)]$, we obtain the 'dispersion' equation

$$q_\parallel^2=\frac{1}{2}\left(\frac{ck_2}{n_o\omega\tau_p^2}+bq_\perp^2\right)q_\perp^2. \tag{32}$$

The first and second terms in parentheses in (32) correspond to nonlinear refraction and diffraction, respectively. For $q_\parallel^2>0$, the soliton solution is stable. One can see from (32) that if the carrier frequency ω of the ordinary component of the soliton lies in the region of the positive GVD, i.e., $\omega=\omega_+$ [see (23)], the soliton is always stable with respect to small-scale self-focusing.

For the carrier frequency of the ordinary component of the soliton $\omega=\omega_-$, we have $k_2<0$ (see above). In this case, $q_\parallel^2>0$ if

$$q_\perp^2>q_{\perp m}^2=\frac{c|k_2|}{n_o\omega b\tau_p^2}=\frac{6n|k_2|\Omega}{c\tau_p^2}. \tag{33}$$

Thus, diffraction can compensate for self-focusing if the size $l\sim1/q_\perp$ of small-scale perturbations is sufficiently small ($l<l_m=1/q_{\perp m}$).

According to [26], we establish the integral criterion for soliton self-focusing. One can easily show that system (1) is equivalent to the nonlinear Schrödinger equation with the five-order nonlinearity,

$$i\frac{\partial Q}{\partial z}=-g\Delta_\perp Q+\frac{\eta}{g}|Q|^4Q, \tag{34}$$

where $g=\pm(b/2)^{1/2}$, $\eta=ck_2/(4n_o\omega)$, and the complex function Q is related to ρ and Φ by the expression

$$Q=\sqrt{\rho}\exp\left[\frac{i}{2g}\left(\frac{ck_2\Omega^2}{n_o\omega}z+\Phi\right)\right]. \tag{35}$$

As a result, the soliton-stability problem is reduced to the analysis of stability of a nonlinear spatial beam described by expression (34).

To Eqn (34), the 'Hamiltonian'

$$H=\int(g^2|\nabla_\perp Q|^2+\frac{\eta}{3}|Q|^6)dS \tag{36}$$

corresponds. In addition, the law of conservation of quantity

$$N=\int|Q|^2dS$$

follows from (34). Here, integration is performed over the entire transverse plane.

Let us define the square of the transverse radius of the soliton as the second-order moment [26]

$$R^2 = \frac{1}{N}\int r^2|Q|^2 \mathrm{d}S, \tag{37}$$

where r is the radial component of the cylindrical coordinate system. By using the method described in [26], we find from (37) and (34)

$$\frac{\mathrm{d}R^2}{\mathrm{d}z} = \frac{2}{N}\int \boldsymbol{j}\boldsymbol{r}\,\mathrm{d}S, \tag{38}$$

where $\boldsymbol{j} = \mathrm{i}g[Q(\nabla_\perp Q^*) - Q^*(\nabla_\perp Q)] = \rho\nabla_\perp\varphi$.

By differentiating (38) once more, we obtain

$$\frac{\mathrm{d}^2R^2}{\mathrm{d}z^2} = \frac{16}{N}H = \mathrm{const}. \tag{39}$$

We assume that the wave fronts of the ordinary component at the input to the medium for $z = 0$ are plane, i.e., $\nabla_\perp\varphi_{|z=0} = 0$. From this, by using (38) and the expression of $\boldsymbol{j}$, we find that $(\mathrm{d}R^2/\mathrm{d}z)_{|z=0} = 0$. Then, after integrating (39), we obtain

$$R^2 = R_0^2 + \frac{8H}{N}z^2, \tag{40}$$

where R_0 is the input radius of the soliton. It follows from (40) that for $H > 0$, the soliton is defocused and for $H < 0$ it undergoes self-focusing.

By using this integral criterion, we estimate the critical power P_{cs} of the soliton above which the self-focusing regime should be observed. It is clear that this will occur inevitably if the integrand in (36) is always negative (this requires the fulfilment of the condition $\eta \sim k_2 < 0$, giving $\omega = \omega_-$). By assuming that $|\nabla_\perp Q|^2 \sim |Q|^2/R^2 = \rho/R^2$ and expressing then ρ in terms of ψ_{m}, we find

$$\psi_{\mathrm{m}}^2R^2 > \frac{c^3|k_2|}{8\pi^2\chi_{\mathrm{eo}}^2\omega_-}.$$

Taking into account that the intensity of the ordinary component is $I_{\mathrm{o}} = c\psi_{\mathrm{m}}^2/4\pi n_{\mathrm{o}}$, we obtain its power,

$$P_{\mathrm{os}} > P_{\mathrm{cs}} \cong \frac{c^4|k_2|}{32\pi^2 n_{\mathrm{o}}\chi_{\mathrm{eo}}^2\omega_-}. \tag{41}$$

Assuming that $|k_2| \sim 1/c\omega_-$, $\chi_{\mathrm{eo}} \sim 10^{-9}$ esu, $n_{\mathrm{o}} \sim 2$ and $\omega_- \cong 3\times10^{14}$ s^{-1}, we obtain $P_{\mathrm{cs}} \sim 10^{10}$ W. For the intensity $I_{\mathrm{o}} \sim 10^{11}$ W cm^{-2}, this power corresponds to a rather broad soliton aperture, of the order of a few millimetres. For apertures 10–100 μm, the critical power is inaccessible and therefore the soliton propagation in the self-channeling regime can be expected.

4. Conclusions

We have proposed a new mechanism of the continuous frequency down-conversion of an optical pulse propagating in a uniaxial crystal. The ordinary component of the pulse undergoes the red shift, by generating synchronously a video pulse of the extraordinary wave due to nonlinearity. For this process to be efficient, the condition of degenerate three-wave mixing or the Zakharov–Benney resonance expressed formally by relations (19) and (22) should be fulfilled. It is shown that the Stokes self-scattering due to the ZBR can occur only in crystals with the positive birefringence. In addition, the angle φ between the pulse propagation direction and the optical axis of the crystal is bounded below, while the carrier frequencies ω_+ and ω_- of the pulse [see (23)] continuously depend on φ. Therefore, to obtain the efficient frequency conversion, a certain value of φ should be selected for each value of the input carrier frequency.

For $\omega = \omega_+$, a down-converted soliton propagates in the defocusing regime, which can be observed in the visible range of input frequencies. If $\omega = \omega_-$, the spectrum of the ordinary component belongs to the negative GVD on the near-IR region. As a result, the Stokes self-scattering due to the ZDR can be observed in the soliton self-channeling regime.

An important difference of the Stokes self-scattering mechanism considered in the paper from SRS is that self-scattering is accompanied by the saturation of the red shift, whose value is proportional to the input pulse intensity, whereas no saturation is observed in the SRS soliton regime and the Stokes shift increases proportionally both to the input intensity and the path propagated by the pulse [2–5]. In addition, in our case not only the down-conversion of the carrier frequency of the ordinary component occurs but also a video pulse of the extraordinary wave is generated, which is also a specific feature of the ZBR mechanism in a uniaxial crystal.

Acknowledgements. This work was supported by the Russian Foundation for Basic Research (Grant No 05-02-16422a).

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