

Spectral study of resonance absorption upon three-wave mixing of femtosecond pulses in nonlinear crystals

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Abstract. The shortened equations are obtained for the spectral components of parametrically interacting pulses in nonlinear-optical crystals, which take into account the dispersion of all the parameters of the medium. The range of applicability of the model is indicated. The effect of resonance absorption on the characteristics of femtosecond pulses is considered.

Keywords: parametric interaction, femtosecond pulses, resonance absorption.

1. Introduction

One of the urgent problems of nonlinear optics is the frequency conversion of femtosecond pulses preserving their temporal parameters [1–4]. A special feature of such processes is that the interacting pulses have the spectral width from tens to hundreds of nanometres. To solve the frequency-conversion problem, a model is needed which would adequately describe the action of all mechanisms affecting the frequency-conversion process. This process is determined by the spectral dependences of the following parameters of a medium: the dispersion of the refractive index $n(\omega)$, the dispersion of the nonlinear coupling coefficient $\gamma(\omega)$ caused by the intrinsic linear frequency dependence and the dispersion of the active nonlinearity coefficient $d_{\text{eff}}(\omega)$ [$\gamma(\omega) \sim \omega d_{\text{eff}}(\omega)$], and the dispersion of the nonlinear absorption coefficient $\alpha(\omega)$.

The frequency-conversion problem is commonly based on the numerical solution of the coupled system of linear wave equations describing variations in the fields of the interacting pulses [5–23]. As a rule, only the first order of the refractive-index dispersion, which is responsible for the group dispersion of the pulses, is taken into account. In [5–15], the frequency-conversion processes were considered within the framework of this mechanism only. The frequency conversion of ultrashort pulses was studied [16–22] taking into account also the second-order dispersion responsible for the dispersion spread of pulses.

The influence of the dispersion of the nonlinear coupling coefficient $\gamma(\omega)$ was studied in [23] within the framework of a temporal model in the absence of the refractive index and absorption dispersion. The effect of the intrinsic dispersion of the nonlinear coupling coefficient $\gamma(\omega)$ was considered, while the nonlinear dependence $d_{\text{eff}}(\omega)$ observed at the boundaries of the transparency region [24] was neglected.

Although the influence of linear-absorption mechanisms is obvious, this absorption was ignored in the nonlinear-optical frequency conversion of broadband radiation. Note that a nonlinear dependence of the absorption coefficient $\alpha(\omega)$ at the boundary of the transparency region and the presence of resonance absorption peaks should be taken into account in the analysis of frequency conversion. For example, the resonance absorption peaks are observed in the region between 2.5 and 5 μm in the KTP, KTA, RTP, and RTA crystals [25, 26], which are efficiently used for conversion of radiation from a femtosecond Cr:forsterite laser to the mid-IR region [27]. The effect of such absorption peaks on the laser-radiation parameters has not been studied so far.

The nonlinearity of processes requires the simultaneous consideration of all the limiting mechanisms. The use of a temporal model for solving the frequency-conversion problem taking into account the dispersion of different orders for all parameters of the medium leads to the appearance of many terms in equations. In this case, the dispersion dependences of the parameters of the medium are approximated by polynomial functions. The appearance of new terms in the equations describing frequency conversion corresponds to the consideration of new terms in the polynomial expansion of dispersion dependences. Because all the basic properties of crystals are determined by dispersion dependences, it is reasonable to solve the frequency-conversion problem in the spectral region, i.e., to use a spectral model. In this case, all the dispersion orders for the parameters of the medium are automatically taken into account, and the experimental dispersion dependences of these parameters can be used without their approximation.

Spectral models were used to analyse the formation of spatial parameters of radiation upon frequency conversion [28–38] and to study processes based on the Kerr nonlinearity (see, for example, [6, 39]). The spectral method was also used to analyse the formation of temporal parameters [40–43]. In this case, as a rule, the shortened wave equations, in which certain restrictions had been made, were used as the initial equations. In [40], the wave detuning between all the spectral components was neglected, which

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Received 7 June 2004; revision received 23 December 2004
Kvantovaya Elektronika 35 (3) 257–262 (2005)
Translated by M.N. Sapozhnikov

actually takes place even in case of the exact phase matching and substantially affects the frequency-conversion process. Note also that in all these papers, absorption and all the more absorption dispersion were neglected.

The problems of frequency conversion for obtaining the spectral distribution of the harmonic used for pulse-duration measurements (for example, by the method of frequency-resolved optical gating (FROG) [44]) were considered separately. These problems, which can be also solved by the spectral method, do not require the high conversion efficiency, and are solved in the fixed-field approximation by neglecting the most of limiting mechanisms (see, for example, [45]).

The aim of this paper is to develop a spectral model for solving the problems of frequency conversion for femtosecond pulses under the conditions of a strong energy exchange taking into account the dispersion of all basic parameters of a medium (refractive index, absorption coefficient, effective nonlinearity and nonlinear coupling coefficients) and to analyse the influence of resonance absorption on the characteristics of laser radiation.

2. Description of the spectral model

Let us derive the equations of our spectral model as in case of the temporal model [5], by solving, however, the wave equation not for the fields of the interacting waves but for their spectra. The wave equation in the plane-wave approximation

$$\frac{\partial^2 E_i}{\partial z^2} = \mu_0 \frac{\partial^2 D_i}{\partial t^2}, \quad i = x, y, z \quad (1)$$

is taken as the initial one. Here, E_i is the field strength;

$$D_i = \varepsilon_0 E_i + \varepsilon_0 P_i = \varepsilon_0 E_i + \varepsilon_0 (P_i^L + P_i^{NL}) \quad (2)$$

is the electric induction; μ_0 and ε_0 are the permeability and permittivity; and P_i is the component of polarisability of the medium.

In case of highly efficient frequency conversion of femtosecond pulses, it is important to preserve the temporal profile of the pulse. For this reason, the interaction regime is selected in which the radiation intensity is lower than the self-action intensity. Otherwise, the self-action effects caused by the third-order nonlinearity of the medium will lead to the deformation of the temporal profile and spectrum of generated radiation [7]. In this paper, the self-action effects were neglected, assuming that the radiation intensity is lower than the self-action intensity.

The linear polarisability spectrum $\mathcal{P}_i^L(\omega)$ has the form [46]

$$\mathcal{P}_i^L(\omega) = \frac{1}{2} \sum_k \chi_{ik}(\omega) E_k(\omega), \quad i, k = x, y, z, \quad (3)$$

where $\chi_{ik}(\omega)$ is the dielectric susceptibility tensor.

Taking into account that the spectrum of the total electric field is the sum of spectra of interacting pulses, the field spectrum can be written in the form

$$E(\omega) = \sum_m \frac{1}{2} [\mathcal{E}^{(m)}(\omega) + \mathcal{E}^{(m)*}(\omega)], \quad m = 1, 2, 3, \quad (4)$$

where $\mathcal{E}^{(m)}$ are the components of the complex spectrum of the m th pulse with the central frequency ω_m . The expression for the nonlinear polarisability spectrum will contain a few terms corresponding to all the types of three-wave interaction: generation of the sum, difference, and combination frequencies for all combinations of the interacting fields. For example, for the component of the nonlinear polarisability spectrum upon generation of the wave with the central frequency ω_3 , which corresponds to the generation of the sum frequency of pulses with central frequencies ω_1 and ω_2 , we obtain the expression

$$\begin{aligned} \mathcal{P}_i^{(3)NL}(\omega) &= \frac{1}{2} \sum_k \sum_j \int_{-\infty}^{\infty} \chi_{ikj}^{(2)}(\omega; \omega', \omega - \omega') \\ &\times [\mathcal{E}_k^{(1)}(\omega') \mathcal{E}_j^{(2)}(\omega - \omega') + \mathcal{E}_k^{(2)}(\omega') \mathcal{E}_j^{(1)}(\omega - \omega')] d\omega', \\ &i, k, j = x, y, z, \end{aligned} \quad (5)$$

and for the modulus of the polarisability vector the expression

$$\begin{aligned} \mathcal{P}^{(3)NL}(\omega) &= \int_{-\infty}^{\infty} d_{\text{eff}}(\omega; \omega', \omega - \omega') \mathcal{E}^{(1)}(\omega') \\ &\times \mathcal{E}^{(2)}(\omega - \omega') d\omega', \end{aligned} \quad (6)$$

where d_{eff} is the effective nonlinearity coefficient for the selected type of interaction (see, for example, [47]). In (6), the frequency-interchange property of the tensor $\chi_{ikj}^{(2)}(\omega''; \omega', \omega'' - \omega') = \chi_{ijk}^{(2)}(\omega''; \omega'' - \omega', \omega')$ is taken into account. For the spectrum of nonlinear polarisability responsible for generation of radiation with central frequencies ω_1 and ω_2 , the expressions are similar.

Let us introduce the notation $\tilde{E}^{(m)}(\omega) = \mathcal{E}^{(m)}(\omega - \omega_m)$, $\tilde{D}^{(m)}(\omega) = \mathcal{D}^{(m)}(\omega - \omega_m)$ (where $\mathcal{D}^{(m)}$ are the components of the complex induction spectrum for the m th pulse). In the temporal representation, this is equivalent to the expression $\tilde{E}^{(m)}(t, z) = \mathcal{E}^{(m)}(t, z) \exp(i\omega_m t)$. By using (2), (3), (6), the shift theorem $[\tilde{E}(\omega, z) \equiv \mathcal{E}(\omega - \omega_0, z)]$, and the condition $\omega_3 = \omega_2 + \omega_1$, we obtain

$$\begin{aligned} \tilde{D}^{(3)}(\omega) &= \varepsilon_0 \tilde{E}^{(3)}(\omega) + \varepsilon_0 \chi(\omega + \omega_3) \tilde{E}^{(3)}(\omega) \\ &+ \varepsilon_0 \int_{-\infty}^{\infty} d_{\text{eff}}(\omega + \omega_3; \omega' + \omega_1, \omega - \omega' + \omega_2) \\ &\times \tilde{E}^{(1)}(\omega') \tilde{E}^{(2)}(\omega - \omega') d\omega'. \end{aligned} \quad (7)$$

Such a representation of the spectra is convenient for numerical simulations because in this case all the spectra are represented relative to zero. For brevity, we introduce the notation

$$d_{\text{eff}}(\omega + \omega_3; \omega' + \omega_1, \omega - \omega' + \omega_2) = d_{\text{eff}}(\omega, \omega').$$

We will seek the solution for $\tilde{E}^{(m)}(\omega, z)$ in the form $\tilde{E}^{(m)}(\omega, z) = A^{(m)}(\omega, z) \exp[-ik_m(\omega + \omega_m)z]$. Taking into

account that $k(\omega) = n(\omega)\omega/c$ and substituting (7) into (1), we obtain

$$\begin{aligned} & \left\{ \frac{\partial^2 A^{(3)}(\omega, z)}{\partial z^2} + \left[-2ik_3(\omega + \omega_3) \right] \frac{\partial A^{(3)}(\omega, z)}{\partial z} \right\} \\ & \times \exp[-ik_3(\omega + \omega_3)z] = -\frac{(\omega + \omega_3)^2}{c^2} \int_{-\infty}^{\infty} d\omega' d_{\text{eff}}(\omega, \omega') \\ & \times A^{(1)}(\omega') \exp[-ik_1(\omega' + \omega_1)z] \\ & \times A^{(2)}(\omega - \omega') \exp[-ik_2(\omega - \omega' + \omega_2)z] d\omega'. \quad (8) \end{aligned}$$

A change in the amplitude of a plane wave propagating in quadratically nonlinear medium is caused by the dispersion of parameters of the medium, the absorption and nonlinear interaction of fields in the medium. A change in the amplitude of the field spectrum is caused only by absorption and nonlinear effects because the dispersion of parameters affects only the phase of the spectrum. A degree of influence of absorption and nonlinear interaction on the amplitude of the field spectrum will be the same as for the filed amplitude. Therefore, if we assume that the method of slowly varying amplitudes is applied to the field, it will be all the more applied to the field spectrum. Let us assume that

$$\left| \frac{\partial^2 A(\omega, z)}{\partial z^2} \right| \ll k \left| \frac{\partial A(\omega, z)}{\partial z} \right|, \quad (9)$$

then, expression (8) will take the form

$$\begin{aligned} \frac{\partial A^{(3)}(\omega, z)}{\partial z} &= -i \int_{-\infty}^{\infty} \frac{d_{\text{eff}}(\omega, \omega')(\omega + \omega_3)^2}{2c^2 k_3(\omega + \omega_3)} A^{(1)}(\omega') \\ & \times A^{(2)}(\omega - \omega') \exp \{ i [k_3(\omega' + \omega_3) - k_1(\omega - \omega' + \omega_1) \\ & - k_2(\omega + \omega_2)] z \} d\omega'. \end{aligned}$$

In the general case, the absorption coefficient can be taken into account by introducing the complex propagation constant. An alternative variant is the factorisation of the initial wave equation with the separation of a term describing attenuation (see, for example, [5]). In passing to the spectra of interacting pulses, the contribution of linear absorption is described by the term $[-\alpha(\omega + \omega_m)/2]A^{(m)}(\omega)$. The coefficient

$$\gamma_m(\omega) = \frac{d_{\text{eff}}(\omega, \omega')(\omega + \omega_m)^2}{2k_m(\omega + \omega_m)c^2} \quad (10)$$

is called the nonlinear coupling coefficient and characterises the degree of nonlinear interaction between the spectral components of interacting pulses. The shortened equations for these components have the form

$$\frac{\partial A^{(3)}(\omega, z)}{\partial z} = -i \int_{-\infty}^{\infty} d\omega' \gamma_3(\omega) A^{(1)}(\omega') A^{(2)}(\omega - \omega') \times$$

$$\begin{aligned} & \times \exp \{ i [k_3(\omega + \omega_3) - k_1(\omega' + \omega_1) - k_2(\omega - \omega' + \omega_2)] z \} \\ & - \frac{1}{2} \alpha(\omega + \omega_3) A^{(3)}(\omega), \\ & \frac{\partial A^{(2)}(\omega, z)}{\partial z} = -i \int_{-\infty}^{\infty} d\omega' \gamma_2(\omega) A^{(3)}(\omega') A^{(1)*}(\omega - \omega') \\ & \times \exp \{ -i [k_3(\omega' + \omega_3) - k_1(\omega - \omega' + \omega_1) - k_2(\omega + \omega_2)] z \} \\ & - \frac{1}{2} \alpha(\omega + \omega_2) A^{(2)}(\omega), \quad (11) \\ & \frac{\partial A^{(1)}(\omega, z)}{\partial z} = -i \int_{-\infty}^{\infty} d\omega' \gamma_1(\omega) A^{(3)}(\omega') A^{(2)*}(\omega - \omega') \\ & \times \exp \{ -i [k_3(\omega' + \omega_3) - k_2(\omega - \omega' + \omega_2) - k_1(\omega + \omega_1)] z \} \\ & - \frac{1}{2} \alpha(\omega + \omega_1) A^{(1)}(\omega). \end{aligned}$$

It can be easily shown that the Parseval equality is not fulfilled for individual spectral components (11), but it is satisfied for the integrated spectra of the interacting pulses. This reflects the fact that all the spectral components of radiation pulses incident on the crystal contribute to each spectral component at the sum frequency. Therefore, the formation of spectral components is determined by the total amplitude and phase distributions of the interacting pulses. The presence of the wave detuning for the interacting spectral components in the general case leads to the redistribution of the power density in the sum-frequency spectrum and reduces the conversion efficiency.

The system of equations (11) describes frequency conversion taking completely into account all the dispersion mechanisms mentioned above. The equations were derived by using the assumptions and approximations determining the conditions of applicability of our spectral model:

(i) The effects of nonlinear self-action are small, i.e., the B-integral (nonlinear phase shift) does not exceed π . Therefore, the pump intensity is limited by the value

$$I_{\text{max}} \leq I_{\text{sa}} = \frac{\lambda}{2\tilde{n}_2 n_0 L_{\text{cr}}}$$

at which the self-action of radiation in the crystal develops. For example, for an LBO crystal of length 5 mm, we have $I_{\text{sa}} = 400 \text{ GW cm}^{-2}$ ($\lambda = 1.25 \text{ }\mu\text{m}$, $n_0 = 1.6$, $\tilde{n}_2 = 2 \times 10^{-16} \text{ cm}^2 \text{ W}^{-1}$ [4]).

(ii) The influence of spatial effects is small (geometrical optics approximation), i.e., the walk-off of extraordinary waves is negligibly small, which takes place if $\tan \beta < r_0/L_{\text{cr}}$ (where β is the walk-off angle and r_0 is the beam radius), and the diffraction length satisfies the relation $L_{\text{dif}} = r_0^2/\lambda \ll L_{\text{cr}}$.

For the typical walk-off angle $\beta < 2^\circ$, the crystal length $L_{\text{cr}} \sim 1 \text{ mm}$, and the wavelength $\lambda \sim 1 \text{ }\mu\text{m}$, the radius r_0 should be smaller than $30 \text{ }\mu\text{m}$. For $r_0 = 30 \text{ }\mu\text{m}$, the pulse energy $E = 100 \text{ }\mu\text{J}$ and duration $\tau = 100 \text{ fs}$, the radiation intensity is $I = 25 \text{ TW cm}^{-2}$. Under these conditions, the

spatial structure of the beam can be described by the method of geometrical optics [46].

3. Analysis of the effect of absorption dispersion on the parameters of generated radiation

As mentioned above, analysis of the dispersion and nonlinear properties of nonlinear-optical crystals has shown that the KTP, KTA, RTP, and RTA crystals can be efficiently used for parametric generation of femtosecond pulses in the range from 2.5 to 5 μm upon pumping by a 1.25- μm femtosecond Cr:forsterite laser [27]. However, these crystals have a few absorption peaks in the range between 2.5 and 4.5 μm [25, 26]. The transmission spectra of these crystals are presented in Fig. 1. For comparison, the spectrum of a 200-fs pulse is also shown in Fig. 1.

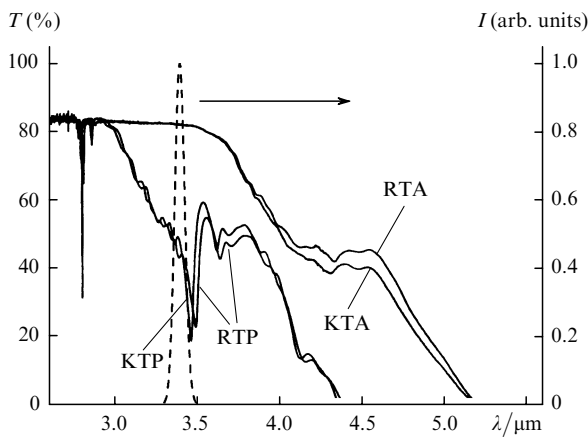


Figure 1. Transmission spectra T of the KTP ($L_{\text{cr}} = 10$ mm), KTA (10 mm), RTP (11 mm), and RTA (11 mm) crystals [26] and the spectrum $I(\lambda)$ of a 200-fs, 3.4- μm pulse.

Consider the influence of these absorption peaks on the characteristics of parametrically generated IR radiation in the range 2.6–4 μm in a KTP crystal with the help of spectral model (11). Let us perform calculations taking dispersion into account or neglecting it. The numerical calculation was performed using the following parameters of pump and injection radiation and the crystal: the pump radiation: $\lambda = 1.25$ μm , $\tau = 200$ fs, $I = 100$ GW cm^{-2} ; the injected radiation: $\lambda = 1.818$ – 2.407 μm , $\tau = 1$ ps, $I = 10$ MW cm^{-2} (the delay time of the pump radiation with respect to the injection radiation was 300 fs); the KTP crystal: $L_{\text{cr}} = 4$ mm, $\varphi = 90^\circ$, $\theta = 50.7^\circ$.

Tuning of the injected radiation jointly with the corresponding rotation of the crystal allows the tuning of generated radiation. The presence of injected radiation during optical parametric oscillation provides an increase in the frequency-conversion efficiency [48].

Figure 2 shows the spectrum and time envelope of the generated 2.8- μm IR radiation. The hole in the spectrum is caused by resonance absorption at this wavelength and results in the appearance of two new maxima on the time envelope of the pulse – at the left and right from the pulse maximum. Let us define the radiation contrast as the ratio of the peak intensity I_0 to the prepulse intensity I_1 . Figure 3 shows the wavelength dependences of the energy, duration, and contrast of generated IR radiation. The calculations were performed by neglecting absorption dispersion and

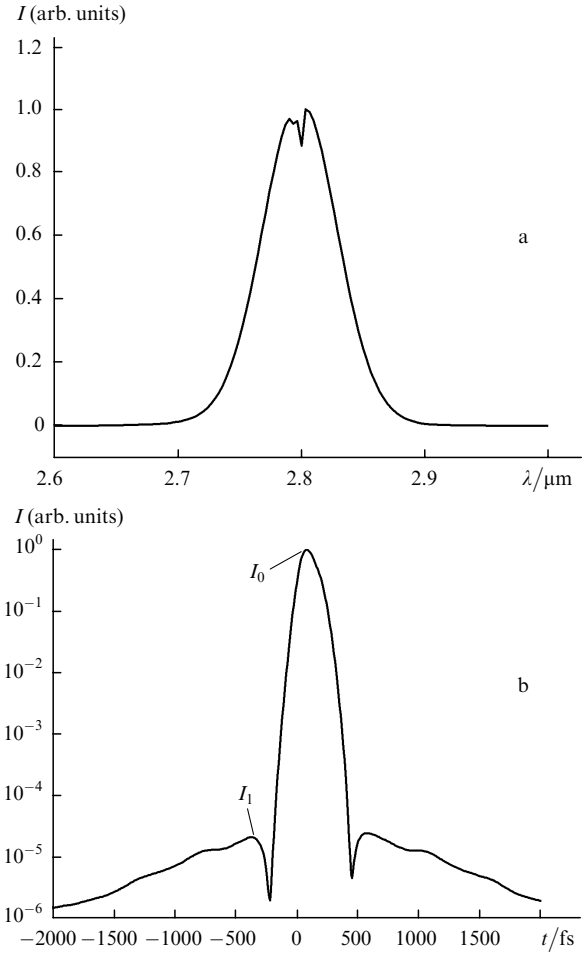


Figure 2. Spectrum $I(\lambda)$ (a) and the time envelope $I(t)$ (b) of the IR pulse generated in the KTP crystal ($\lambda = 2.8$ μm) (I_0 and I_1 are the intensities of the pulse and prepulse, respectively).

also taking into account absorption distributed over the crystal length and localised absorption. In the calculation neglecting dispersion, spectrally uniform absorption equal to the absorption coefficient at the central wavelength was considered. In the calculation with localised absorption, the attenuation having dispersion was taken into account only at the output face of the crystal.

One can see from Fig. 3a that, when the spectral width of the absorption peak is much smaller than the spectral width of the IR pulse, the absorption dispersion should be taken into account. The difference between the results of calculations performed by considering the absorption dispersion or neglecting it amounts to 13%. Note that, despite its large amplitude (40%), the absorption peak at 2.8 μm affects neither the IR-pulse energy nor its duration because of a small width of the peak. The 3.45- μm absorption peak (Fig. 1) also does not change noticeably the IR-pulse energy and only slightly changes its duration (no more than by 5 fs).

The absorption peaks of the crystal do not change significantly the IR-pulse duration and energy; however, as one can see from Fig. 3c, they considerably deteriorate the contrast of the IR pulse compared to that for radiation propagating through a medium without absorption dispersion. The results of calculation for the case of distributed absorption, when the radiation contrast is determined by the combined action of resonance absorption and nonlinear

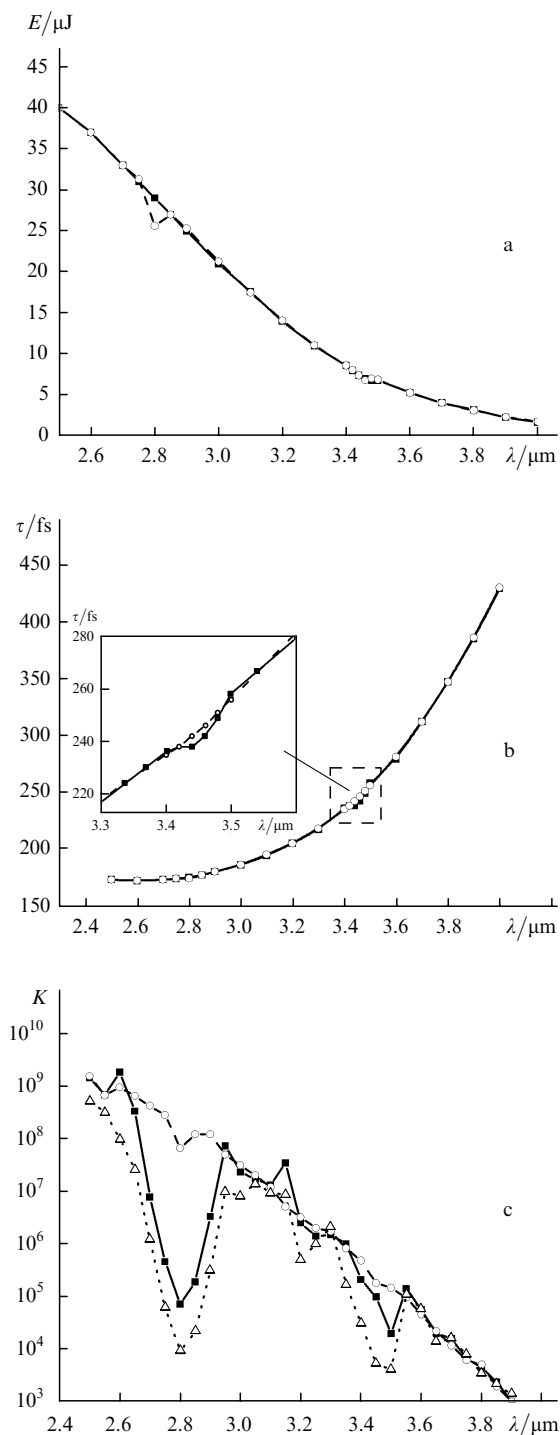


Figure 3. Wavelength dependences of the energy E (a), duration τ (b), and contrast K (c) of the IR pulse generated in the KTP crystal ($\lambda = 2.8 \mu\text{m}$) obtained by neglecting absorption dispersion (\circ) and taking it into account medium with distributed (\blacksquare) and localised (\triangle) absorption.

conversion, differ by 10 times from those obtained for the case of localised absorption, when the contrast is determined only by resonance absorption. Therefore, we can conclude that nonlinear frequency conversion prevents the contrast lowering caused by resonance absorption compared to the contrast lowering in the absence of absorption.

The calculations show that the resonance change in the absorption coefficient does not cause a substantial change in the energy and duration of generated radiation. However, in

the general case the resonance absorption is characterised not only by a change in the absorption coefficient but also in the refractive index. Obviously, a resonance change in the refractive index will cause variations that are similar to the group dispersion and dispersion spread of pulses, which are inherent in the refractive-index dispersion. The influence of the resonance absorption peaks can be taken into account more accurately by using the experimental dependences of not only the absorption coefficient but the refractive index as well.

4. Conclusions

The spectral model of the three-wave mixing of pulses has been proposed which takes into account the dispersion of the main parameters of the medium such as the refractive index, the effective nonlinearity coefficient, the absorption coefficient, and the nonlinear coupling coefficient. The range of applicability of the model is indicated. Analysis of the effect of resonance absorption on optical parametric oscillation in the KTP crystal has shown that, first, the resonance absorption substantially impairs the radiation contrast compared to that of radiation propagating through a medium without absorption dispersion and, second, the nonlinear frequency conversion prevents the contrast reduction caused by absorption compared to that in the case of localised absorption. To describe the frequency-conversion process near resonance absorption peaks more accurately, it is necessary to solve this problem by the spectral method by using the experimental spectral dependences of the refractive index and absorption coefficient.

Acknowledgements. The author is indebted to the late V.I. Pryalkin for his participation in these studies and also thanks V.M. Gordienko, V.A. Trofimov, and S.G. Grechin for useful discussions of the paper. This work was supported by the Russian Foundation for Basic Research (Grant No. 03-02-16973).

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