

Compression of Laser Pulses in the Interaction of Counterpropagating Waves in a Medium with an Inertial Cubic Nonlinearity

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Abstract—The theory of the interaction of high-power counterpropagating laser pulses in a transparent medium with an inertial cubic nonlinearity is developed. An effect of temporal compression of pulses due to nonstationary cross-phase modulation, which results in energy exchange between the interacting pulses, is predicted. It is demonstrated that the direction of energy exchange is independent of the sign of the nonlinearity parameter of a medium. For input pulses with different intensities, energy exchange can occur in the anomalous direction—from a weak pulse to a strong pulse. The considered specific features of the nonstationary interaction of counterpropagating pulses are manifested in schemes of multiwave mixing employed for the phase conjugation of short laser pulses.

1. INTRODUCTION

Since high-power ultrashort pulses of coherent radiation are used for the investigation of ultrafast relaxation processes in matter, for the study of the interaction of atoms and molecules with superstrong fields, etc., the development and creation of optical systems for the compression (temporal focusing) of laser pulses is of considerable importance for the solution of fundamental problems and various applications [1].

Optical compressors based on the compression of phase-modulated laser pulses in dispersive media [2] have been thoroughly studied and have found wide acceptance in practice. The phasing of the spectral components of a laser pulse drastically shortens the pulse and increases its peak intensity. However, in practice, it is rather difficult to implement the limiting regime of pulse compression. Therefore, much effort is being made in this field to remove aberrations arising in modulators and compressors proper, to improve energy characteristics and the stability of compressed pulses, to control the shape of such pulses, etc. [1, 3].

In this paper, we propose a comparatively simple method of temporal compression of laser pulses using the interaction of laser pulses in a medium with an inertial cubic nonlinearity. The cross-action of laser pulses in a medium with an inertial nonlinear response gives rise to nonstationary energy exchange between the pulses [4, 5]. Such cross-action of the colliding pulses results in temporal compression of pulses accompanied by an increase in the peak power. Compression can be achieved in the absence of the initial phase modulation either for identical or for different (in the power, shape, and duration) laser pulses.

Based on the linearized wave equations that describe the interaction of counterpropagating stepwise laser pulses, we found an explicit form of the function that governs the spatial and temporal evolution of the envelopes of laser pulses. We determined conditions of optimal compression of pulses for various lengths of the medium L and different relaxation times t_0 of the nonlinear response. The derived set of wave equations is analyzed for the general case by means of computer simulation. We investigated the distortion of the shape of Gaussian pulses involved in multiple interaction (collisions) in counterpropagating waves in a layer of a medium with an inertial nonlinear response. We determined the ultimate degree of pulse compression as a function of the number of passes through a layer of a nonlinear medium and the ratio of the time L/v required for a laser pulse to pass through the medium to the relaxation time t_0 of the nonlinear response (i.e., the parameter $\mu = L/vt_0$, where v is the speed of light in the medium).

2. THE BASIC EQUATIONS

If the nonlinear addition δn to the refractive index n_0 of a transparent medium is related to inertial effects (e.g., high-frequency Kerr effect), then the dynamic equation for δn is written as [1]

$$t_0(d\delta n/dt) + \delta n = n_2|E|^2, \quad (1)$$

where n_2 is a constant that characterizes the nonlinear addition and E is the amplitude of the light field. For pulses propagating in opposite directions (along the z -axis),

$$E = (E_+e^{ikz} + E_-e^{-ikz})e^{-i\omega t}, \quad (2)$$

the solution to equation (1) is written in terms of the integral

$$\delta n = \frac{n_2}{t_0} \int_{-\infty}^t (|E_+|^2 + |E_-|^2 + 2\operatorname{Re} E_+ E_-^* e^{2ikz}) e^{-\frac{t-t'}{t_0}} dt'. \quad (3)$$

Here, $E_{\pm}(z, t)$ are the slowly varying amplitudes of the pulses and ω and $k = (\omega/c)n_0$ are the mean frequency and the wave number, respectively. The substitution of (2) and (3) into the wave equation yields a set of equations for slowly varying complex amplitudes of pulses [6],

$$\pm \frac{\partial E_{\pm}}{\partial z} + \frac{1}{v} \frac{\partial E_{\pm}}{\partial t} = i \frac{\gamma_0}{t_0} \left(E_{\pm} \int_{-\infty}^t (|E_+|^2 + |E_-|^2) e^{-\frac{t-t'}{t_0}} dt' + E_{\mp} \int_{-\infty}^t E_{\mp}^* E_{\pm} e^{-\frac{t-t'}{t_0}} dt' \right), \quad (4)$$

where $v = c/n_0$ and $\gamma_0 = kn_2/n_0$.

The boundary conditions of the problem are written as

$$E_+(0, t) = E_{+0}(t), \quad E_-(L, t) = E_{-0}(t). \quad (5)$$

To analyze the set (4), it is convenient to use the substitution $E_{\pm} = A_{\pm} \exp(i\varphi_{\pm})$ and to consider equations for real amplitudes $A_{\pm}(z, t)$ and phases $\varphi_{\pm}(z, t)$:

$$\pm \frac{\partial A_{\pm}}{\partial z} + \frac{1}{v} \frac{\partial A_{\pm}}{\partial t} = \pm \frac{\gamma_0}{t_0} A_{\mp} \int_{-\infty}^t A_+ A_- \sin U(t, t') e^{-\frac{t-t'}{t_0}} dt', \quad (6)$$

$$\pm \frac{\partial \varphi_{\pm}}{\partial z} + \frac{1}{v} \frac{\partial \varphi_{\pm}}{\partial t} = \frac{\gamma_0}{t_0} \left\{ \int_{-\infty}^t (A_+^2 + A_-^2) e^{-\frac{t-t'}{t_0}} dt' + \frac{A_{\mp 0}}{A_{\pm 0}} \int_{-\infty}^t A_+ A_- \cos U(t, t') e^{-\frac{t-t'}{t_0}} dt' \right\}, \quad (7)$$

where $U(t, t') = \Delta\varphi(z, t) - \Delta\varphi(z, t')$; $\Delta\varphi(z, t) = \varphi_+(z, t) - \varphi_-(z, t)$; and, correspondingly, $E_{\pm 0} = A_{\pm 0} \exp(i\varphi_{\pm 0})$. For $t_0 \rightarrow 0$, the right-hand sides of equations (6) vanish. In other words, the amplitudes of laser pulses propagating in a medium with an instantaneous nonlinear response remain unchanged. In the case of an inertial nonlinear response (when $t_0 \neq 0$), the right-hand sides of equations (6) differ from zero, and the amplitudes of interacting pulses that propagate in the considered medium are subjected to the corresponding changes [7]. Note that, in the case of an instantaneous nonlinear response, the set of equations (4) has an exact analytical solution, which was obtained by S.A. Akhmanov and his collaborators [8].

3. SOLUTION OF THE LINEARIZED EQUATIONS

We will analytically study the specific features of the evolution of the amplitudes of counterpropagating pulses in a medium with an inertial nonlinear response using equations (6) and (7) linearized near the values corresponding to the following boundary conditions of the problem:

$$A_{+0}(t) = A_{-0}(t) = A_0 = \text{const}, \quad (8)$$

for $t \geq 0$, $\varphi_{\pm 0}(t) = 0$.

Setting

$$A_+(z, t) = A_{+0}(t - z/v) + a_+(z, t), \quad |a_+| \ll A_0, \quad (9)$$

$$A_-(z, t) = A_{-0}(t - (L - z)/v) + a_-(z, t), \quad |a_-| \ll A_0, \quad (10)$$

and assuming that $|U(t, t')| \ll 1$, we can apply (6) and (7) to derive the following equations for a_{\pm} and φ_{\pm} :

$$\pm \frac{\partial a_{\pm}}{\partial z} + \frac{1}{v} \frac{\partial a_{\pm}}{\partial t} = \pm \frac{\gamma_0}{t_0} A_{\mp 0} \int_0^t A_{+0} A_{-0} U(t, t') e^{-\frac{t-t'}{t_0}} dt', \quad (11)$$

$$\pm \frac{\partial \varphi_{\pm}}{\partial z} + \frac{1}{v} \frac{\partial \varphi_{\pm}}{\partial t} = \frac{\gamma_0}{t_0} \left\{ \int_0^t (A_{+0}^2 + A_{-0}^2) e^{-\frac{t-t'}{t_0}} dt' + \frac{A_{\mp 0}}{A_{\pm 0}} \int_0^t A_{+0} A_{-0} e^{-\frac{t-t'}{t_0}} dt' \right\}. \quad (12)$$

From the symmetry of equations (11) and (12) and boundary conditions (8), we find that $a_+(L, t) = a_-(0, t) \equiv a_0(t)$ and $\varphi_+(L, t) = \varphi_-(0, t) \equiv \varphi_0(t)$.

Linearized equations (11) and (12) govern the transfer of amplitudes and phases of the counterpropagating pulses and form an uncoupled set of inhomogeneous equations (with known source terms in the right-hand sides). The solutions to these equations are written in terms of the integrals [9]

$$H_{\pm}(z, t) = \pm \int_{z_0}^z F_{\pm} \left(z', t \mp \frac{z - z'}{v} \right) dz', \quad (13)$$

where $H_{\pm} = \{a_{\pm}, \varphi_{\pm}\}$, F_{\pm} are the functions of the sources, $z_0 = 0$ for H_+ , and $z_0 = L$ for H_- . Integration limits within various regions of z are determined by the space-time dependence of the amplitudes $A_{\pm 0}(z, t)$ of the counterpropagating pulses [9]. Using integrals (13) and taking into account formulas (8)–(10) and the relation $\varphi_+(z, t) = \varphi_-(L - z, t) = \varphi(z, t)$, we can represent the

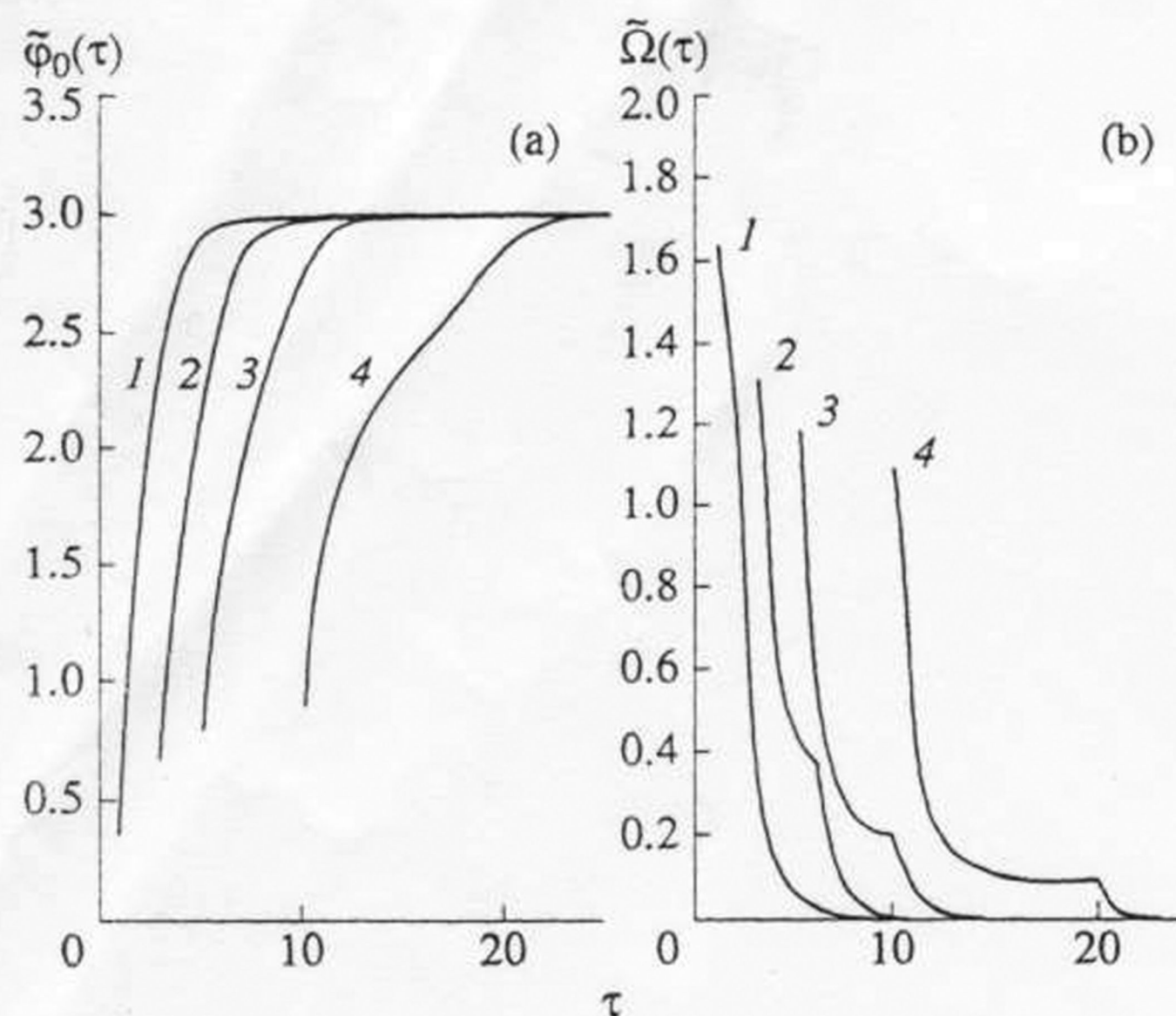


Fig. 1. Evolution of (a) the normalized phase $\tilde{\varphi}_0(\tau) = \varphi_0(\tau)/\gamma_0 A_0^2 L$ and (b) the normalized deviation of the carrier frequency $\tilde{\Omega}(\tau) = \Omega(\tau)/\gamma_0 A_0^2 L$ within the envelope of laser pulses for $\gamma_0 > 0$ and various values of the parameter μ : (1) $\mu = 0.4$, (2) 3, (3) 5, and (4) 10.

solution to equations (12) in the following form:

$$\varphi(z, t) = \gamma_0 A_0^2 \begin{cases} z(1 - e^{-\xi}), & \text{for } (0 \leq z \leq vt, \quad 0 \leq t \leq L/2v) \\ (0 \leq z \leq L - vt, \quad L/2v \leq t \leq L/v) \\ z(1 - e^{-\xi}) + (z - L + vt) - vt_0(1 - e^{-\eta_-}), & \text{for } (L - vt \leq z \leq vt, \quad L/2v \leq t \leq L/v) \\ (0 \leq z \leq L, \quad L/v \leq t \leq (L+z)/v) \\ z(1 - e^{-\xi}) + 2z - vt_0(e^{-\eta_-} - e^{-\eta_+}), & \text{for } (0 \leq z \leq L, \quad t \geq (L+z)/v), \end{cases} \quad (14)$$

where $\xi = (t - z/v)/t_0 \geq 0$ and $\eta_{\pm} = (t - (L \mp z)/v)/t_0 \geq 0$. As can be seen from (14), for $t \gg t_0, L/v$, the stationary phase incursion of the pulses at the output of the medium is $\varphi_{st} = 3\gamma_0 A_0^2 L$.

Using (14), we can determine the dimensionless frequency deviation $\Omega(\tau) = (\omega - \bar{\omega})t_0 = d\varphi_0(\tau)/d\tau$ due to cross-phase modulation within the envelope of the output pulses,

$$\Omega(\tau) = \gamma_0 A_0^2 L \begin{cases} \frac{1}{\mu} + \left(e^{\mu} - \frac{1}{\mu}\right)e^{-\tau}, & \text{for } \mu \leq \tau \leq 2\mu \\ \left(e^{\mu} + \frac{1}{\mu}e^{2\mu} - \frac{1}{\mu}\right)e^{-\tau}, & \text{for } \tau \geq 2\mu, \end{cases} \quad (15)$$

where $\bar{\omega}(\tau)$ is the instantaneous frequency of the output pulses and $\tau = t/t_0$. Obviously, the sign of the frequency

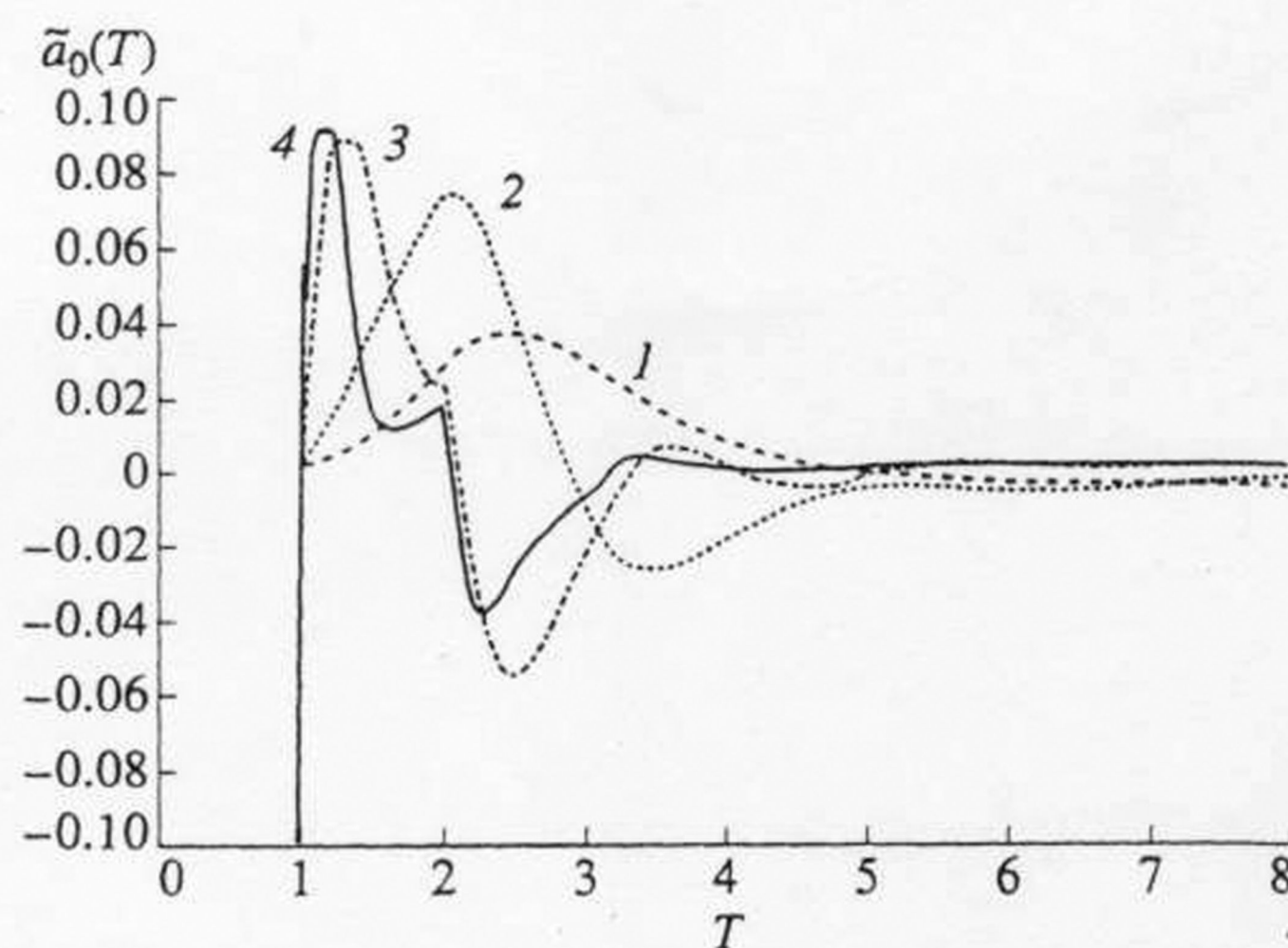


Fig. 2. Time dependences of the normalized function $\tilde{a}_0(T) = \sqrt{\gamma_0 L} a_0(T)$ for $\gamma_0 A_0^2 L = 1$ and various values of the parameter μ : (1) $\mu = 0.4$, (2) 1, (3) 5, and (4) 10.

deviation is determined by the sign of the nonlinearity parameter of the medium, $\gamma_0 \sim n_2$. Figure 1 presents the plots of the normalized functions $\varphi_0(\tau)$ and $\Omega(\tau)$. As can be seen from these plots, the frequency deviation reaches its maximum on the leading edges of the pulses. Note that, for large μ ($\mu \gg 1$), we have $1/\mu \leq |\Omega| \leq 1$ within the interval $\mu \leq \tau \leq 2\mu$. Estimating the relevant parameters as $t_0 = 10^{-12}$ s, $\gamma_0 A_0^2 L = 10^{-1}$, and $\mu = 10^2$, we can use (15) to find that the Stokes shift (or the anti-Stokes shift if $\gamma_0 < 0$) of the frequency ω of the output pulses falls within the interval $10^{11} \geq |\omega - \bar{\omega}| \geq 10^9$ s $^{-1}$.

Using formula (13) for the general solution to the wave equations with allowance for (14) and performing straightforward but sufficiently labor-consuming calculations, we can represent the expression for the function $a_0(\tau)$ in the form

$$a_0(\tau) = A_0(\gamma_0 A_0^2 L)^2 \sum_{\kappa, \sigma} d_{\kappa} \tau^{\sigma} e^{-G_{\kappa\sigma} \tau}, \quad (16)$$

where d_{κ} and $G_{\kappa\sigma}$ are constant coefficients.¹

Figure 2 displays the time dependence of the normalized function $a_0(T)$ ($T = tv/L$) calculated in accordance with (16) for various values of the parameter μ in the case when $\gamma_0 > 0$. As can be seen from these plots, for small μ ($\mu < 1$), the function $a_0(T)$ has a form of a dispersion curve. As the parameter μ increases, which corresponds to a decrease in the relaxation time t_0 of the nonlinear response, the maximum of the function $a_0(T)$ is shifted toward smaller T and is sharpened ($a_0^{\max} \approx 8 \times 10^{-2}$).

¹ Since we are limited in space here, rather cumbersome explicit formulas for the coefficients d_{κ} and $G_{\kappa\sigma}$ will be presented elsewhere.

Subsequently, the function $a_0(T)$ becomes more complex and features additional maxima and minima (see the curves for $\mu = 5$ and 10). The performed calculations demonstrate that, within the range $10 < \mu < 40$, the function $a_0(T)$ only slightly changes its form, and $a_0(T) \rightarrow 0$ with a further growth in the parameter μ . The minimum duration of the first (main) spike at the half-maximum level is approximately 0.3μ .

The evolution of the amplitudes of the counterpropagating pulses governed by the function $a_0(T)$ is due to nonstationary energy exchange [4, 5, 10], which gives rise to energy transfer to the leading edges of the pulses through the cross-action of light waves regardless of the sign of the nonlinearity parameter of the medium, $n_2 \sim \gamma_0$. It is well known that energy exchange through frequency-degenerate two-wave mixing is forbidden in a medium with an instantaneous nonlinear response [11]. In the case of an inertial nonlinear response of a medium, cross-phase modulation changes the instantaneous frequency of pulses within pulse envelopes, and, in fact, frequency-nondegenerate mixing is implemented in the interaction of counterpropagating pulses, so that energy exchange is no longer forbidden. Specifically, the gain of a weak wave ($|E_-(\omega_-)| \ll |E_+(\omega_+)|$), which characterizes the efficiency of energy exchange in frequency-nondegenerate mixing, can be represented in the form [12]

$$g(\omega_-) \sim \gamma_0 |E_+|^2 \frac{\omega_+ - \omega_-}{t_0^{-2} + (\omega_+ - \omega_-)^2}. \quad (17)$$

Obviously, for $\gamma_0 > 0$, amplification [$g(\omega_-) > 0$] is implemented in the Stokes spectral region. For $\gamma_0 < 0$, amplification occurs in the anti-Stokes region. As can be seen from (15), the following inequalities are satisfied in the case under consideration regardless of the sign of n_2 :

$$g(\bar{\omega}_+) \sim \gamma_0 |E_-|^2 (\bar{\omega}_- - \bar{\omega}_+) \begin{cases} \geq 0 & \text{for } L/2 \leq z \leq vt \\ \leq 0 & \text{for } 0 \leq z \leq L/2, \end{cases} \quad (18)$$

$$g(\bar{\omega}_-) \sim \gamma_0 |E_+|^2 (\bar{\omega}_+ - \bar{\omega}_-) \begin{cases} \geq 0 & \text{for } L - vt \leq z \leq L/2 \\ \leq 0 & \text{for } L/2 \leq z \leq L, \end{cases} \quad (19)$$

where $\bar{\omega}_\pm(z, t)$ are the instantaneous frequencies of the forward, E_+ , and backward, E_- , pulses. Inequalities (18) and (19) are written for the region where $L/2v \leq t \leq L/v$. Hence, nonstationary energy exchange results in the amplification of the leading edges of the pulses, where frequency conversion occurs with a higher efficiency (see Fig. 1). Predominant amplification of the leading edges of the pulses is naturally accompanied by the depletion of energy in the remaining parts of pulse envelopes. Obviously, for symmetric boundary conditions (8), the integral pulse energies remain unchanged, and the net effect of nonstationary energy exchange is

reduced to the redistribution of energy within the pulses, which is manifested in the distortion of pulse envelopes. As the interaction of pulses approaches the stationary regime, $\Omega \rightarrow 0$. Correspondingly, energy exchange ceases, and pulse envelopes become stable. We can demonstrate that, in the case of amplitude asymmetry of boundary conditions (8), $A_{+0} \neq A_{-0}$, energy exchange can occur in the anomalous direction in the considered geometry: from a weak pulse to a strong pulse [13].

Note that, in terms of dynamic holography [4], nonstationary energy exchange can be interpreted as phase mismatch between the light-induced grating of the refractive index of the medium and the interference pattern of the counterpropagating pulses. Such mismatch is characteristic of frequency-degenerate two-wave mixing in media with an inertial nonlinear response [4].² Self-diffraction of radiation from a phase-mismatched grating in a medium with an inertial response gives rise to energy exchange between the writing pulses.

4. RESULTS OF NUMERICAL SIMULATION

For the purposes of numerical simulation, it is convenient to write the set of equations (4) in terms of dimensionless variables $x = z/L$ and T :

$$\begin{aligned} & \pm \frac{\partial \mathcal{E}_\pm}{\partial x} + \frac{\partial \mathcal{E}_\pm}{\partial T} \\ & = i\mu(\text{sgn } \gamma_0) e^{-\mu T} \left(\mathcal{E}_\pm \int_{-\infty}^T (|\mathcal{E}_+|^2 + |\mathcal{E}_-|^2) e^{\mu T'} dT' \right. \\ & \quad \left. + \mathcal{E}_\mp \int_{-\infty}^T \mathcal{E}_\mp^* \mathcal{E}_\pm e^{\mu T'} dT' \right), \end{aligned} \quad (20)$$

where $\mathcal{E}_\pm = \sqrt{|\gamma_0| L} E_\pm$.

We solved the set of equations (20) with the use of numerical methods for rectangular and Gaussian input pulses. The results of numerical simulation (the output intensities of the pulses) are presented in Figs. 3–5.

As can be seen from Figs. 3 and 4, in the considered range of parameters, a growth in μ leads to an increase in the peak intensities of the output pulses. Correspondingly, the pulse duration decreases (the total energy of the pulses remains unchanged within an accuracy of 4%). The peak intensities of the output pulses increase approximately by a factor of 1.5. We can demonstrate that, similarly to the linear case (see Section 3), the influence of nonstationary energy exchange becomes weaker for $\mu > 40$. As a result, the intensities of the output pulses virtually coincide with the intensities of the input pulses within this region. An analogous evolution

² The light-induced grating is automatically mismatched from the interference pattern of the writing field in media with a nonlocal nonlinear response [4].

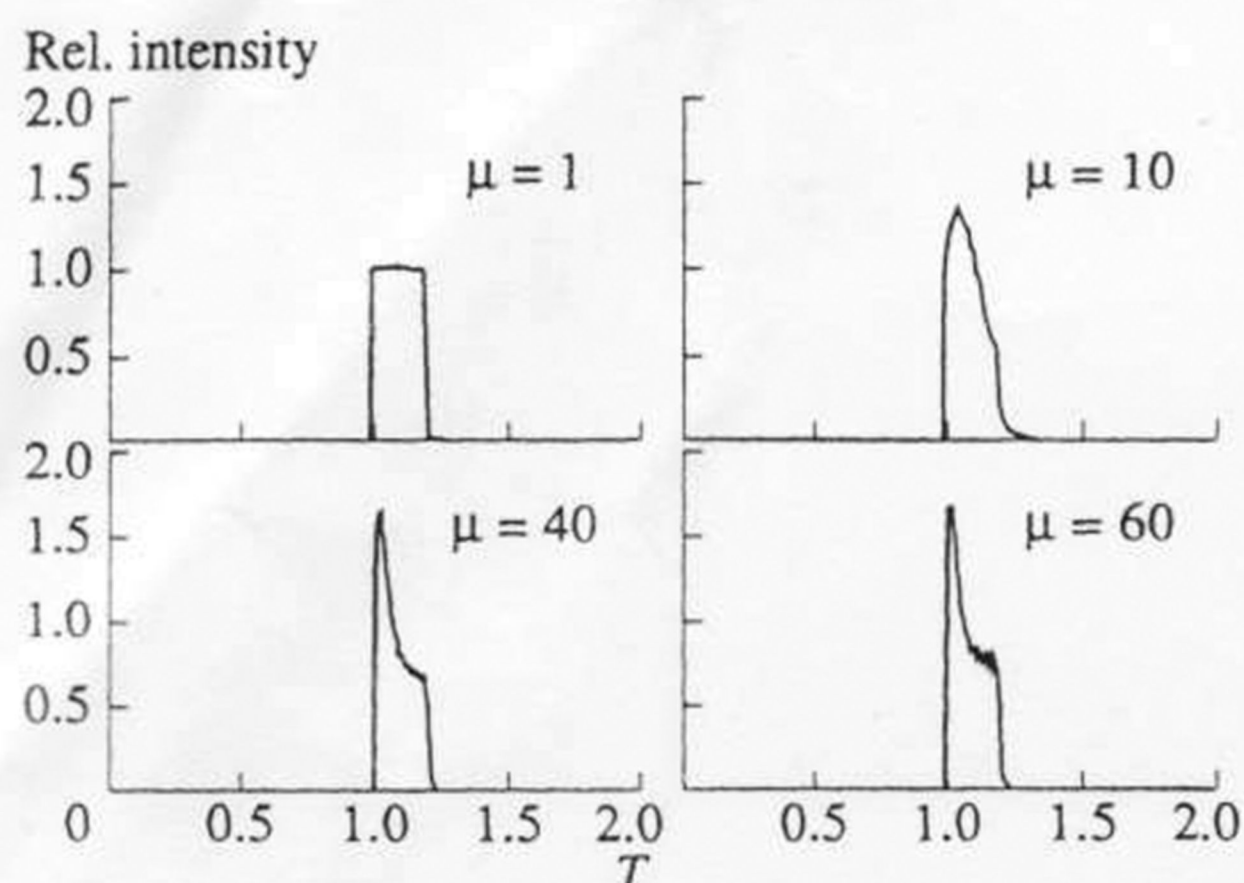


Fig. 3. Envelopes of the relative intensities of the output pulses for various values of the parameter μ in the case of rectangular input pulses $|\mathcal{E}_{\pm 0}(T)|^2 = 10$ within the range $0 \leq T \leq T_p$, where $T_p = 0.2$ is the normalized pulse duration.

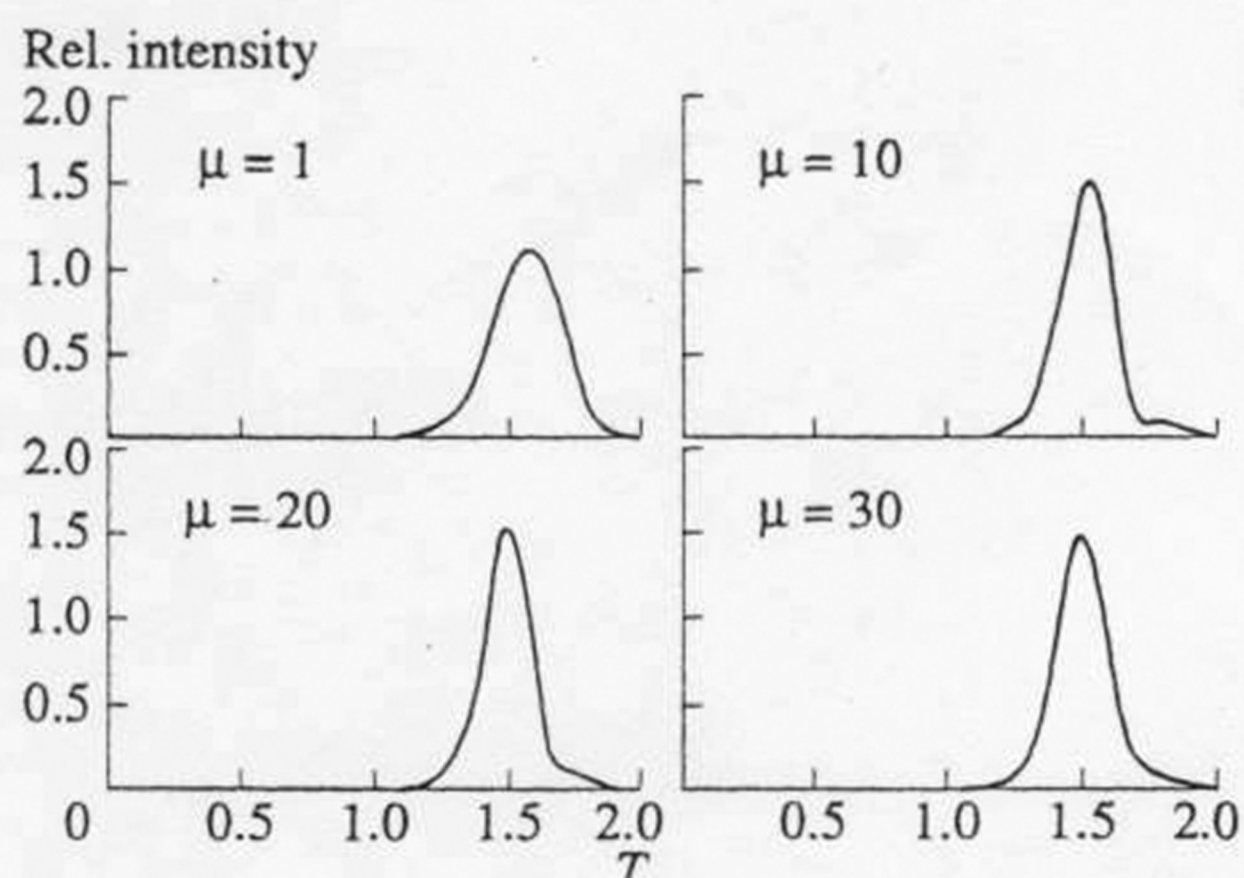


Fig. 4. Envelopes of the relative intensities of the output pulses for various values of the parameter μ in the case when $|\mathcal{E}_{\pm 0}(T)|^2 = |\mathcal{E}_0|^2 \exp[-(T - 3T_p)^2/T_p^2]$ with $|\mathcal{E}_0|^2 = 10$ and $T_p = 0.2$.

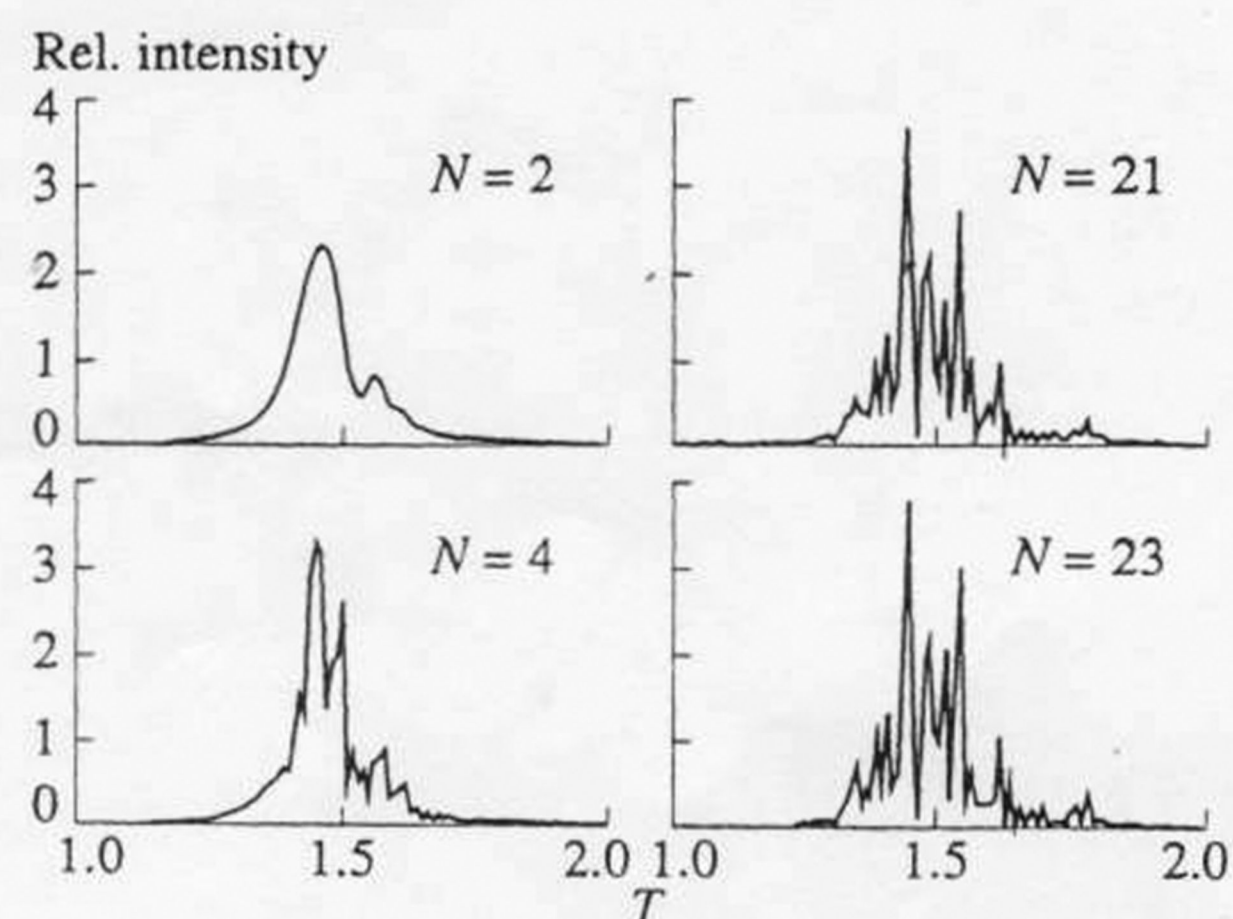


Fig. 5. Envelopes of the relative intensities of the output pulses for various N with $\mu = 30$. Parameters of the input pulses are the same as in Fig. 4.

of the pulses also occurs when the input intensities of the pulses are not equal to each other. If the input pulses are switched on at different moments of time, then the pulse is applied to a medium earlier and, correspondingly, covers a greater distance in the medium before the collision point features a more considerable variation in the shape.

To achieve the ultimate temporal compression of pulses, one can use, for example, a ring cavity, which makes it possible to implement multiple collisions of pulses in a nonlinear medium. Numerical simulations demonstrated that, after an appropriate number of collisions, the output pulses stabilize their shape and display virtually no variations later on.

Figure 5 shows the envelopes of the intensities of the output pulses for different numbers N of pulse collisions in the nonlinear medium in the case when the input pulses have Gaussian shapes and equal intensities. To conserve the symmetry, the procedure of numerical simulations implied the elimination of errors that occurred in the calculation of the output intensities of the forward and backward pulses. For the chosen parameters, the shape of the output pulses became stable after $N > 20$ pulse collisions, whereupon the output pulses acquired a complex temporal structure with a clearly pronounced narrow peak, whose duration was approximately an order of magnitude less than the duration of the input pulses.

5. CONCLUSION

In this paper, we performed theoretical analysis of the interaction of high-power counterpropagating laser pulses in a transparent medium with an inertial cubic nonlinearity. Based on this investigation, we predicted the effect of pulse compression due to nonstationary cross-phase modulation, which gives rise to energy exchange between the pulses and, as a consequence, results in the distortion of pulse envelopes. It is demonstrated that the direction of energy exchange is independent of the sign of the nonlinearity parameter n_2 of a medium. For input pulses with different intensities, energy exchange can occur in the anomalous direction—from a weak pulse to a strong pulse. Due to the conservation of the integral radiation energy (in the absence of dissipation), pulse compression is accompanied by the corresponding increase in the peak intensity of the pulses. The considered specific features of the interaction of counterpropagating pulses should be taken into account in various problems of nonlinear optics, including analysis of multiwave mixing for the phase conjugation of short laser pulses.

In conclusion, we should note that, in combination with a nonlinear saturable filter, which makes it possible to improve the contrast of temporal characteristics of radiation, the considered multipass compression scheme may be of practical interest for various problems of laser physics. The effect of anomalous energy

transfer from a weak pulse to a strong pulse can be employed to produce profiled pulses.

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