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NONLINEAR-OPTICAL PHENOMENA

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Influence of the cubic spectral phase of high-power laser pulses on their self-phase modulation

V.N. Ginzburg, A.A. Kochetkov, I.V. Yakovlev, S.Yu. Mironov, A.A. Shaykin, E.A. Khazanov

Abstract. **Spectral broadening of high-power transform-limited laser pulses under self-phase modulation in a medium with cubic nonlinearity is widely used to reduce pulse duration and to increase its power. It is shown that the cubic spectral phase of the initial pulse leads to a qualitatively different broadening of its spectrum: the spectrum has narrow peaks and broadening decreases. However, the use of chirped mirrors allows such pulses to be as effectively compressed as transform-limited pulses.**

Keywords: *femtosecond pulses, high-power lasers, cubic nonlinearity.*

Reducing the duration of high-power femtosecond pulses is limited by both the spectral amplification band and the transmission band of a stretcher–compressor system. In practice, this leads to the fact that the pulse duration at the output of high-power lasers is 25–40 fs for lasers based on Ti:sapphire crystals or parametric amplifiers and hundreds of femtoseconds for Nd:glass lasers [1]. In such lasers, the only way to shorten a pulse is the broadening of its spectrum with the help of self-phase modulation and subsequent compression by correction of the spectral phase [2, 3]. This method has long been used in low-power (with pulse energies of less than 1 mJ) lasers by using either a fibre [4, 5], or a capillary [6, 7], or a bulk medium [8, 9]. In the latter case, the efficiency of the method is limited by the spatial self-phase modulation nonuniformity associated with the bell shape of the beam. The solution to this problem is to use a negative lens as a nonlinear element [10]. For example, Mironov et al. [11] demonstrated the reduction of durations from 40 to 20 fs for a 28-mJ pulse. In petawatt lasers, the pulse energy of which amounts to tens of joules, the method has not been applied until recently due to a lack of glass or crystal elements with an aperture of more than 10 cm and a thickness of less than 1 mm. It is shown in [2, 12] that the use of polymer materials, such as polyethylene terephthalate, can solve this problem. Another important distinctive feature of high-power lasers consists in the following: their pulses are generally not transform-limited due to the stretcher–compressor system nonideality and material dispersion of the active medium, and the problem of spectrum broadening under self-phase modulation has been investigated only for transform-limited pulses, starting with a classical book [13].

V.N. Ginzburg, A.A. Kochetkov, I.V. Yakovlev, S.Yu. Mironov, A.A. Shaykin, E.A. Khazanov Institute of Applied Physics, Russian Academy of Sciences, ul. Ul'yanova 46, 603950 Nizhnii Novgorod, Russia; e-mail: efimkhazanov@gmail.com

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In this report we have shown that self-phase modulation of a pulse with a residual cubic spectral phase has qualitative peculiarities: the spectrum has narrow peaks and the spectral is considerably less broadened. In addition, we have found that in spite of this fact, such pulses can be as effectively compressed as transform-limited pulses by a simplest phase corrector, which introduces only a quadratic spectral phase.

Consider the problem of the spectrum broadening of femtosecond pulses with an initial cubic spectral phase. The electric field envelope $A(t)$ of the pulse and its spectrum $S(\Omega)$ have the form

$$A(t) = \int_{-\infty}^{\infty} S(\Omega)\exp(\mathrm{i}\Omega t)\,\mathrm{d}\Omega,$$
$$S(\Omega) = S_0 \exp\left[-2\ln 2\frac{\Omega^2}{\Omega_{\mathrm{FWHM}}^2} - \mathrm{i}\varphi(\Omega)\right], \tag{1}$$

where S_0 is the amplitude of the spectrum; Ω is the detuning from the centre frequency; Ω_{FWHM} is the spectral full width at half maximum; and $\varphi(\Omega) = \beta\Omega^3/6$ is the cubic phase. The pulse propagation in a nonlinear medium is described by a quasi-optical equation in the second approximation of dispersion theory:

$$\frac{\partial A}{\partial z} + \frac{1}{u}\frac{\partial A}{\partial t} - \mathrm{i}\frac{k_2}{2}\frac{\partial^2 A}{\partial t^2} + \mathrm{i}\gamma|A|^2 A = 0, \tag{2}$$

where $\gamma = (3\pi k_0\chi^{(3)})/(2n_0^2)$; u is the group velocity; z is the longitudinal coordinate; k_2 is the parameter of the group velocity dispersion; n_0 is the linear part of the refractive index; k_0 is the wave vector; and $\chi^{(3)}$ is the nonlinear susceptibility. The effect of the cubic nonlinearity is determined by the B-integral $B = \gamma|A_{\max}(z=0)|^2 L$, where $A_{\max}$ is the maximum value of the field, and L is the length of the medium. Envelopes of a 50-fs transform-limited pulse ($\beta = 0$) and a pulse with β = 15000, 60000 and 110000 fs^3 leading to their stretching by 5%, 25% and 40%, respectively, are shown in Fig. 1a. The spectra of these pulses are the same [curves (*5*) in Figs 1b–d]. Figures 1b–d show the spectra of self-phase modulated pulses at different values of β and B. One can see that the spectrum of a transform-limited pulse broadens significantly greater than the spectra of pulses with cubic phase. Moreover, the characteristic scale of spectral modulation of the latter pulses is much smaller than that of the transform-limited ones. Note that this is true even in the case of a small

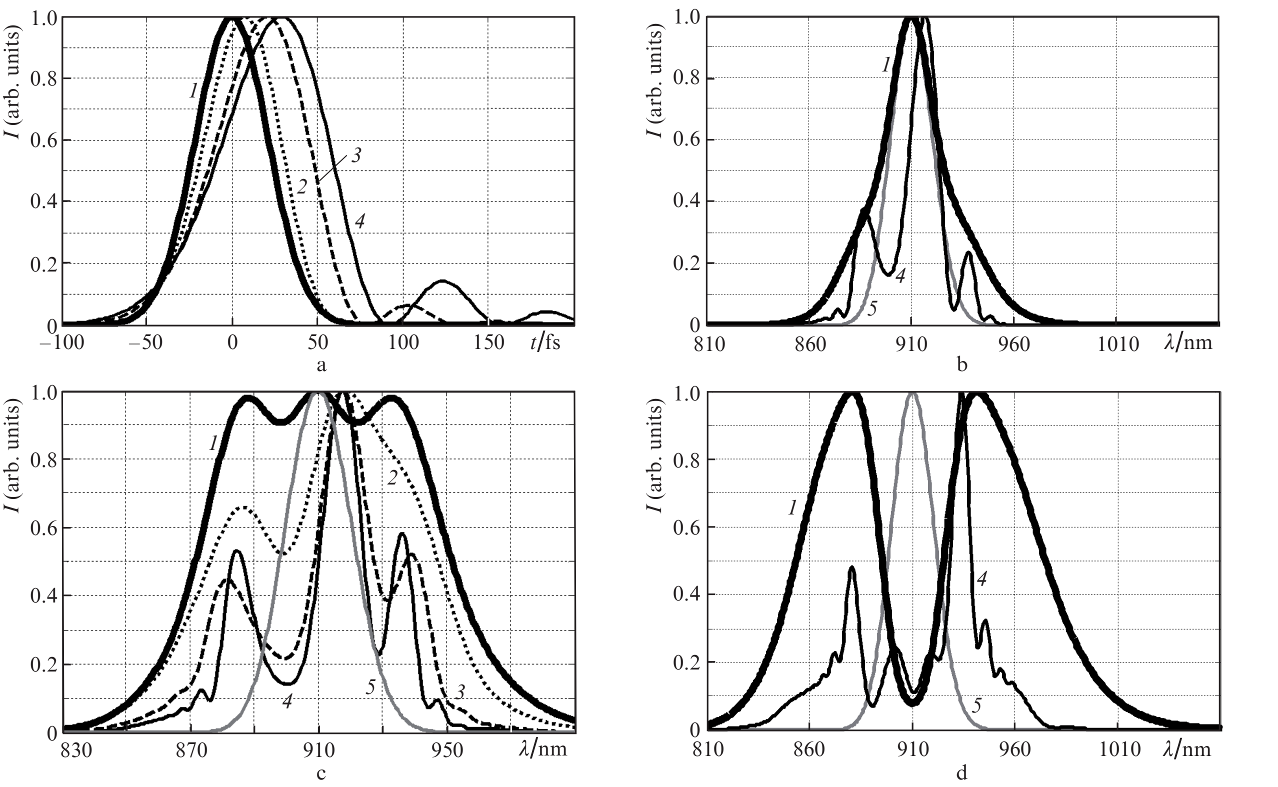

Figure 1. Envelopes of the input pulse (a) and pulse spectra at $B = 2$ (b), 3 (c) and 5 (d) for β = (*1*) 0, (*2*) 15000, (*3*) 60000 and (*4*) 110000 fs^3 and the spectrum of the input pulse (*5*).

phase value (β = 15000 fs^3), at which the pulse envelope distortions are minimal – the stretching is only 5 %, and subpulses are almost not visible. Calculations showed that for a thin nonlinear medium (the thickness of less than 2 mm at a pulse duration of 50 fs) the linear dispersion is not critical and can be neglected at $B \leqslant 5$. In this case, instead of a numerical simulation of equation (2), we can use its analytical solution [13].

Experiments were performed with a 65-fs pulse from the front-end system of the petawatt PEARL laser [14], which is 40 % longer than the duration of a transform-limited pulse. The cubic phase with the parameter β = 110000 fs^3 corresponds to such an increase in the duration. The spectrum of the input pulse is shown in Fig. 2 [curve (*1*)]. The pulse energy was 20 mJ. As a nonlinear medium we used a 1.7-mm-thick glass plate. Figure 2a shows the spectra integrated over the beam cross section at B = 3 and 5. We managed to avoid the breakdown of optical elements at such high values of the B-integral due to the spatial self-filtration [15]. Although the shape of the input pulse spectrum is far from Gaussian, the output pulse spectrum characteristics are the same as those shown in Fig. 1: we observed only a slight broadening of the spectrum and the appearance of narrow peaks. Numerical simulations showed that for a transform-limited pulse with the same spectrum we observe a much greater broadening of the spectrum (Fig. 2b), while the behaviour of the spectrum of the pulse with a cubic spectral phase with β = 110000 fs^3 is qualitatively similar to that observed in the experiment.

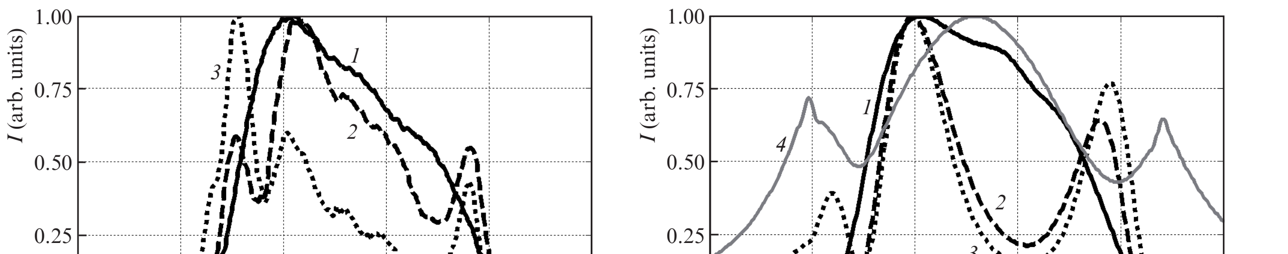

Figure 2. (a) Experimental and (b) theoretical spectra of the input (*1*) pulse and output pulse at B = 3 (*2*) and 5 (*3*), as well as of a transform-limited pulse at B = 5 (*4*).

This strong dependence on the spectral phase of the incident pulse may be used to recover the pulse amplitude and phase from a set of spectrum measurements after self-phase modulation with different values of the B-integral. The development of recovery algorithms is the subject of a separate study.

Consider how effectively phase-modulated pulses can be compressed. We restrict ourselves to the simplest case of a quadratic phase corrector, which can be represented by commercially available chirped mirrors. Mathematically, such a correction is described as follows:

$$A_{\rm c}(t) = F^{-1}\left\{\exp\left(-\frac{{\rm i}\alpha\Omega^2}{2}\right)F[A_{\rm out}(t,z=L)]\right\}, \qquad (3)$$

where F and F^{-1} are the direct and inverse Fourier transforms; $A_{\rm out}(t, z = L)$ is the field at the output from a nonlinear medium; and α is the dispersion parameter of the corrector. Numerically, we found the optimum value $\alpha_{\rm opt}$, at which the maximum amplification of the output pulse power magnification ratio $P_{\rm out}/P_{\rm in}$ is reached. The spectrum of the input pulse was taken the same as in the experiment (Fig. 2a). Results of the optimisation are shown in Fig. 3. Note that in the case of a Gaussian input pulse spectrum, the optimisation results differ slightly.

Figure 3 allows us to draw the following conclusions. Firstly, increasing the power of a cubic phase pulse is almost the same as for a transform-limited pulse. Secondly, in the case of optimal correction, an increase in power with high accuracy depends linearly on the B-integral and is described by the formula

$$P_{\rm out}/P_{\rm in} = 1 + B/2.$$

In particular, it follows from this formula that the power $P_{\rm out}$ in case of two-stage compression at $B = B_{\rm total}/2$ in each stage is greater than in the case of a single-stage compression at $B = B_{\rm total}$. Thirdly, the compression of a cubic phase pulse requires considerably larger dispersion $\alpha_{\rm opt}$ of the phase corrector, which is apparently due to a smaller broadening of its spectrum (Figs 1 and 2). Fourthly, larger values of the B-integral require lower absolute values of $\alpha_{\rm opt}$. If $B \geqslant 3$, one commercially available chirped mirror is sufficient for pulse compression.

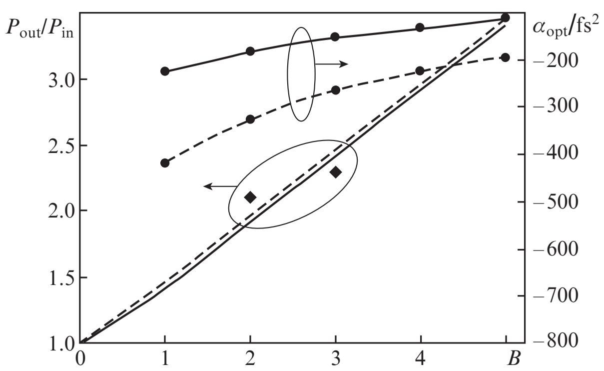

Figure 3. Dependences of the peak pulse power amplification coefficient $P_{\rm out}/P_{\rm in}$ and the corresponding value of $\alpha_{\rm opt}$ on the B-integral for a transform-limited pulse (solid curves) and a pulse with β = 110000 fs^3 (dashed curves). Diamonds show the results of experiments.

Calculations also showed that when α deviates from $\alpha_{\rm opt}$, the value of $P_{\rm out}/P_{\rm in}$ slowly decreases. For example, if $B = 3$, then $P_{\rm out}/P_{\rm in}$ decreases by less than 10% with a change of α from −230 to −100 fs^2 for a transform-limited pulse and from −400 to −170 fs^2 for a pulse with β = 110000 fs^3. Therefore, even a significant error in the calculation and fabrication of a chirped mirror will not lead to a significant reduction in the output pulse power.

Our preliminary experiments on the phase correction have demonstrated good agreement with theory. Figure 3 shows two experimental points for $P_{\rm out}/P_{\rm in}$. A detailed description of these studies will be the subject of a separate publication.

We have observed the effect of a qualitative influence of the cubic spectral phase of a laser pulse on its self-phase modulation in a nonlinear medium at any values of the B-integral. In particular, the pulse spectrum contains narrow peaks and the spectral broadening is substantially less than that for a transform-limited pulse. It is shown theoretically that regardless of the magnitude of the spectral cubic phase of the initial pulse, the use of chirped mirrors, introducing only a quadratic phase, can increase the output pulse power by about 1 + B/2 times.

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