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Influence of predelay between interacting picosecond pulses on the nonlinear optical processes of frequency conversion

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Abstract. The efficiency of second-harmonic generation and the degree of compression of picosecond pulses from a neodymium–glass laser were enhanced by a delay between the ordinary and extraordinary pulses interacting in KDP crystals. The conversion efficiency was increased by a factor of 1.5 (from 33% to 50%) and the second-harmonic pulses were compressed by a factor of 2.4 (from 4.8 to 2 ps).

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The technique of the generation and amplification of "chirped" pulses, which has recently become so popular in the attainment of intensities exceeding 10^{18} W cm^{-2}, makes it possible to implement new regimes for the interaction of high-power laser radiation with matter. The most important parameter here is the contrast of a pulse (i.e. the ratio of the intensity at the pulse peak to the background), which should be $\sim 10^7$ in order to avoid depletion of a pulse caused by the absorption in a plasma created by a prepulse. Frequency conversion of radiation generated by powerful neodymium–glass laser systems combined with second-harmonic generation in nonlinear crystals makes it possible to perform this task. Powerful high-contrast second-harmonic radiation is currently used to generate a dense high-temperature laser plasma [1].

One of the principal aims in conversion to the second harmonic of laser radiation is an increase in the energy and power efficiencies of this nonlinear process. The efficiency of frequency doubling of pulses with durations from fractions of a ps to a few picoseconds is hindered by the influence of the dispersion of the group velocities of the interacting pulses in a nonlinear crystal. For example, when pulses of 2 ps duration are used, the group-velocity dispersion in a KDP crystal encountered in the type-II (oe-e) interaction between ordinary (o) and extraordinary (e) waves of the fundamental radiation and of the second harmonic leads to separation of these waves in space after they have traveled less than 1 cm in a crystal. This limits the energy efficiency of conversion of sub-picosecond pulses to below $\sim 40\%$ for optimal (~ 4 GW cm^{-2}) intensities of the radiation to be converted. At high intensities the second harmonic is converted back to the fundamental frequency and the role of other nonlinear optical processes is enhanced.

The recently proposed [2–4] method for predelay of an e wave relative to an o wave of the fundamental-frequency radiation when it reached a KDP doubler crystal (where the type-II interaction takes place) has made it possible to increase the ultimate energy efficiency of the conversion process and also to compress significantly the converted pulses, increasing greatly the power efficiency [5–7] as well as the contrast of converted pulses [8].

The method is as follows. If e pulses are delayed relative to o pulses by $\sim \tau_f$ (where τ_f is the duration of the pulses), then at some radiation intensities the length of the interaction in a negative nonlinear crystal is doubled and this increases the efficiency of nonlinear optical conversion. The feasibility of simultaneous compression of the converted radiation is related to the following circumstance. If the group velocities are different, then one point on a temporal profile of a second-harmonic pulse interacts with all points of the o and e waves. The energy (75%–80%) [5, 6] and power (240%) conversion efficiencies [7], and a greater than 20-fold pulse compression [9] achieved by this method demonstrate its great promise for solving the tasks outlined above. This method was extended for optical parametric oscillators [10–12]. Recently the feasibility of the compression of tunable pulses by means of sum-frequency generation under special group-velocity mismatch conditions was demonstrated [8]. In this article we demonstrate the use of this method both for an increase of the frequency conversion efficiency in long KDP crystals cut for the type-II phase-matching and for compression of radiation pulses ($\tau_f \sim 5$ ps) of a neodymium–glass laser.

1 Experimental setup

We used a picosecond master oscillator with a Nd:glass active element and negative feedback. Bandwidth-limited pulses were injected into an oscillator–amplifier system [13]. The radiation at the output of the system had the following parameters: pulse energy 10 mJ, duration 4.8 ps, diameter at half-amplitude of the intensity distribution 6 mm, power density up to 8 GW cm^{-2}, and divergence 2×10^{-4} rad.

The apparatus used is shown schematically in Fig. 1. Linearly polarized radiation was passed through a quarter-wave

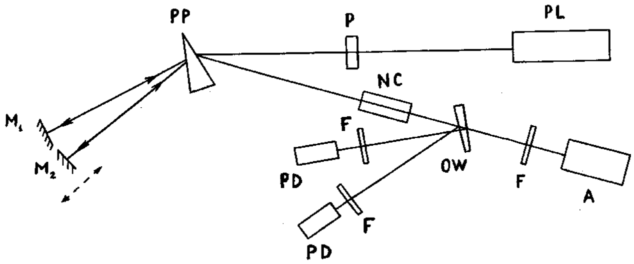

Fig. 1. Schematic diagram of the apparatus: PL–picosecond laser; PP–polarisation prism; M_1, M_2-mirrors; NC–nonlinear crystal; F–filters; A–autocorrelator; PD–photodiodes; P–$\lambda/4$ plate; OW–optical wedge

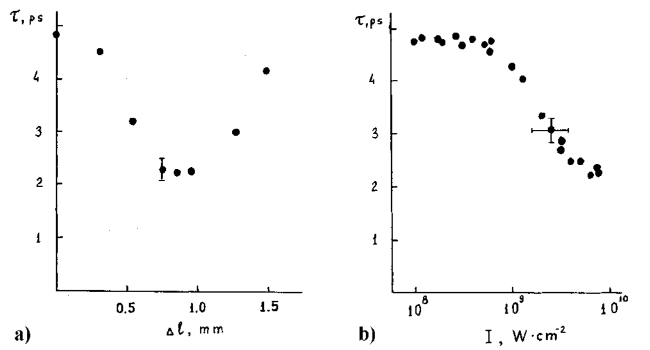

Fig. 2. Dependencies of the duration τ of the converted radiation pulses on the delay between orthogonal polarized pulses obtained for $I = 5\,\mathrm{GW\,cm^{-2}}$ (**a**) and on the intensity of the fundamental (pump) radiation (**b**)

plate (P) and was split by a KDP polarizing prism (PP) into two beams with orthogonal polarization. The beams were reflected by mirrors (M_1 and M_2) and were then combined in the prism so that they passed through a nonlinear crystal (NC) along the same path. The frequency converter was a KDP crystal cut for type-II phase-matching (20 mm × 20 mm × 40 mm). The second harmonic passed through a SZS-21 filter (F) and reached a homemade autocorrelator (A). The radiation energies at the fundamental frequency and at the second harmonic were recorded with calibrated FD-24K photodiodes (PD) and the signals from them were measured with V4-17 pulse voltmeters (not shown in Fig. 1). As was shown in previous works [4, 12] an optimal ratio between intensities of interacted o and e waves $k = (I_o/I_e)$ in the crystal should be close to the ratio of group lengths (L_o/L_e), where $L_o = \tau_f/[1/(v_{e,f}) - 1/(v_{e,2\omega})]$ and $L_e = \tau_f/[1/(v_{o,f}) - 1/(v_{e,2\omega})]$. In our case the ratio $|L_o/L_e|$ was ~ 1.4. We kept the ratio of intensities of o and e waves close to this value in the experiments of pulse compression.

One of the mirrors (M_2) was placed on a stage capable of micro-displacements. This produced a variety of delays between the o and e waves, which interacted in the converter crystal. The autocorrelator was used to determine the duration of the pulses of the interacting fundamental-frequency and second-harmonic radiation, and the time delay between them.

The pulse duration was deduced from noncolinear second-harmonic generation in one shot and also from the spatial distribution of the second-harmonic radiation, which was recorded with a CCD linear array. The information was analyzed on a personal computer and reached a monitor [14]. The temporal characteristics of the second harmonic generated in the KDP crystal (NC) were determined by suitable modifications of the autocorrelator system. In this case, the doubler crystal in the autocorrelator was KDP cut for type-I (oo–e) interaction with $\theta = 85°$; the angle between the interacting beams with the wavelengths $\lambda = 527$ nm was 5°.

2 Experimental results and discussion

In the first series of experiments we determined the dependence of the duration of the second-harmonic pulses on the delay between the o and e waves produced by displacing one of the mirrors (M_2). This gave the results plotted in Fig. 2a. The equality of the optical paths of the o and e waves was deduced from the spatial position of the second harmonic in the autocorrelator, which measured the duration of the fundamental-frequency pulses.

Since the group velocity of the e wave ($\lambda = 1054$ nm) in the KDP crystal exceeded the group velocity of the o wave ($v_{o,f} = 1.966 \times 10^{10}\,\mathrm{cm\,s^{-1}}$, $v_{e,f} = 2.019 \times 10^{10}\,\mathrm{cm\,s^{-1}}$), the delay of the o wave relative to the e wave simply resulted in a steep fall of the conversion efficiency because the waves did not overlap in the mixer crystal. An opposite situation caused compression of the converted radiation pulses. For converted pulses of this duration (4.8 ps), there was an optimal delay between the pulses with orthogonal polarization. A further increase in the delay produced a situation in which the e wave did not catch up with the o wave within the nonlinear crystal. An important condition for efficient compression was intersection inside the crystal of parts of the pulses with the maximum intensities (in the temporal profiles). The duration of the second-harmonic pulses varied from 4.8 ps for the case when the o and e waves coincided at the entry of the nonlinear crystal to 2.2 ps for $\Delta l = 0.9$ mm (Δl is the difference between the optical paths of the e and o waves).

Figure 2b shows the dependency of the duration of the second-harmonic pulses on the intensity ($I = I_o + I_e$) of the fundamental wave ($k = 1.4$) obtained for a fixed delay between the pulses (4 ps). The pulses duration decreased linearly (to 2 ps) with an increase in intensity, this was observed from $0.7\,\mathrm{GW\,cm^{-2}}$. It should be possible to reduce further the duration of the converted pulses and to subsequently increase their intensity, because at the investigated intensities (up to $8\,\mathrm{GW\,cm^{-2}}$) the influence of other limiting factors (Raman scattering, self-focusing, etc.) was slight. The return of the second-harmonic energy to the sub-harmonic could give rise to a structure in the temporal profile of the second-radiation pulses when the intensity is increased.

Figure 3 shows the dependency of the energy-efficiency (η) of conversion to the second harmonic on the delay between the waves. An increase in the delay between the e and o waves increased the conversion efficiency from 33% to 50% (for $k = 1$). However, when the delay was increased still further, the conversion efficiency fell for exactly the same reasons which limited compression in the first series of experiments. A further increase in the conversion efficiency and pulse compression for an optimal delay may be limited by phase modulation of the pulses, which was induced in the preamplifier.

The length of the effective interaction zone (in the crystal) ofthe pulses with orthogonal polarization, $L_{eff} = \tau_f/[1/(v_{o,f}) - 1/(v_{e,f})]$, was 3.6 cm. When KDP crystals with the type-I interaction were used, the influence of the group-

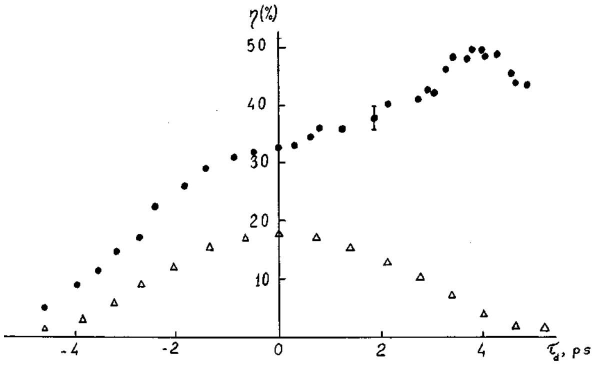

Fig. 3. Dependencies of the efficiency of conversion to the second harmonic on the delay between the e and o waves in KDP crystal with lengths $d = 40$ mm (•) and 4 mm (△)

velocity dispersion could be ignored because the difference between the group-velocities of the fundamental radiation and the second harmonic was slight. However, in view of the much weaker dependency of the conversion efficiency on the angular mismatch and the possibility of using the method of predelay of the interacting pulses–which would make it possible to achieve temporal compression in addition to an increase of η–it would be preferable to employ crystals with the type-II interaction in order to use the second harmonic in the interaction of ultra-strong optical fields with matter. These processes are being currently investigated with high-power neodymium–glass laser systems and amplification of "chirped" pulses.

For comparison, we plotted in Fig. 3 the dependency of the conversion efficiency on the displacement of one of the mirrors (M_2) when a thin (4 mm) KDP crystal with the interaction of the same type was used. The dependency repeated exactly the autocorrelation function of the second harmonic for a step-by-step autocorrelator. This function could be used to determine the duration of the pulses at the fundamental frequency ($\tau_f = \tau_d/\sqrt{2} = 4.8$ ps, assuming a gaussian shape of the pulse).

The dependency of the energy efficiency of the conversion process on the intensity of the fundamental-frequency radiation ($I = I_o + I_e$) is plotted in Fig. 4 for different delays, between the pulses. In the absence of such a delay, the conversion efficiency reached saturation at the intensity $I = 3\,\mathrm{GW\,cm^{-2}}$ ($\eta = 33\%$) and did not vary for a wide range of intensities. Introduction of a 4-ps delay between the pulses had the effect that the maximum conversion efficiency (50%) was achieved at $I = 4\,\mathrm{GW\,cm^{-2}}$; the efficiency then fell to 40% at $8\,\mathrm{GW\,cm^{-2}}$. When the delay was equal to the duration of the fundamental-frequency pulses (4.8 ps), the maximum efficiency (42%) was reached at $I = 5\,\mathrm{GW\,cm^{-2}}$ and fell at higher intensities. The measured contract (ratio of the pulse energy to the background) increased from $10^4 : 1$ to $5 \times 10^5 : 1$ after conversion to the second harmonic in the predelay scheme.

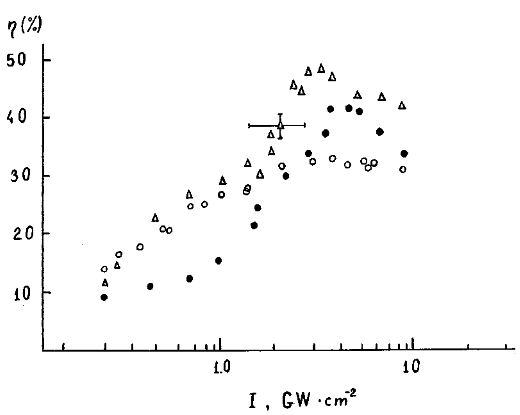

Fig. 4. Dependencies of the energy efficiency of conversion to the second harmonic on the intensity of the fundamental-frequency radiation when the delay between the e and o waves was 4 ps (△), and 4.8 ps (•), and also when there was no delay (○), ($k = 1$)

3 Conclusion

Our experiments demonstrated pulse compression (by a factor of 2.4) and an increase in the conversion efficiency (from 33% to 50%) when the delay between the e and o waves was 3 and 4 ps respectively. The increase in the efficiency of the energy conversion process and the pulse compression were the result of "prolonged" (compared with the usual case) overlap of the e and o waves in the converter crystal. Phase modulation of fundamental waves can play an important role in these processes and restrict the value of one of them.

Our investigations demonstrate the promise of the nonlinear optical methods for the compression of picosecond pulses and an increase of the efficiency of nonlinear processes. Further optimization of the conversion and compression of the pulses, subject to selection of the optimal power density (which may be different for these two processes), can be expected in the future.

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