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Post-compression of a second harmonic pulse: a way to increase the peak power and temporal contrast of ultrahigh-power laser pulses

S Yu Mironov, E A Khazanov

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Contents

Abstract. We discuss the use of second harmonic generation (SHG) and subsequent temporal compression to increase the peak power and improve the temporal contrast ratio of pulses at the output of petawatt and multi-petawatt laser systems. Two ways to apply SHG are considered: directly to ultra-short pulses with central wavelengths of 910 nm and 800 nm, as well as to (sub)-picosecond pulses of neodymium lasers with energies up to 1 kJ. The second way is a new approach to producing sources of ultrahigh-power laser pulses of ultra-short duration in the visible wavelength range and may be used in all modern projects aimed at generating extreme light.

Keywords: second harmonic generation, time contrast, time post-compression

1. Introduction

In modern laser systems with a high peak power (at the TW and PW level) [1–3], as well as in all 100-PW-level projects [4–9], wide use is made of neodymium laser radiation with a central wavelength close to one micron. The pulse energy at the output of such lasers can reach many kJ. A striking example is the French PETAL laser [10, 11]. However, the characteristic spectral width is a few nm, which limits the minimum pulse duration to hundreds of femtoseconds. It is for this reason that in most PW lasers Nd lasers are used as auxiliary radiation, which is employed after frequency doubling to pump laser or parametric amplifiers. This makes it possible to obtain pulses with a duration of tens of fs, but with a significantly lower energy. Solving the problem of reducing the duration from hundreds to tens of and even a few femtoseconds without significant loss of energy will make it possible to generate laser pulses with both high energy and ultrashort duration, which in turn will lead to an increase in peak power and intensity in the region of radiation–target interaction.

Currently, one of the most promising approaches to reducing the duration and increasing the peak power is the method of nonlinear temporal compression (post-compression) [12, 13]. The method involves self-modulation of the phase of a high-power laser pulse as it propagates in a medium with cubic nonlinearity. Such a medium can be thin ($\sim$ mm) dielectric plates made of glass or fused quartz [14], nonlinear crystals [15], or polymers [16–18]. A laser pulse with a duration close to its Fourier limit is directed to them. At the output of the plates, laser radiation is phase-modulated and has a broad spectrum. Subsequent correction of the spectrum phase ensures a reduction in duration. Post-compression can be applied repeatedly [13, 16, 19], with the result that the pulse duration can be reduced to one period of light field oscillations. For pulses with a duration of tens of fs, post-compression was successfully implemented at the output of many high-power laser systems (for example, PEARL [14, 20], ALLS [21], CoReLS [22]), where pulses with a duration of 10 fs or less were obtained. However, for pulses with a duration of hundreds of fs, post-compression was demonstrated only in Ref. [23] for an attenuated replica of the laser beam.

An even more interesting and promising technique for reducing pulse duration is the combination of second harmonic generation and post-compression: phase self-modulation occurs directly in a crystal frequency doubler, and then the spectrum phase is corrected in the second

S Yu Mironov (*), **E A Khazanov** (**)
Federal Research Center A V Gaponov-Grekhov Institute of Applied Physics of the Russian Academy of Sciences (IAP RAS), ul. Ul'yanova 46, 603155 Nizhny Novgorod, Russian Federation
E-mail: (*) sergey.mironov@mail.ru, (**) efimkhazanov@gmail.com

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harmonic pulse [24–26]. With this approach, it becomes possible to convert powerful infrared radiation into the visible range, which would reduce the size of the laser beam at the focus, shorten the pulse length, increase the peak power, increase the at-focus intensity, and improve the temporal contrast. To convert the radiation of ultrahigh-power laser pulses into the second harmonic, use is made, as a rule, of a nonlinear KDP crystal, which is due to the availability of technology for growing large-aperture (20 cm or more) elements of high quality.

In this paper, we consider the features of second harmonic generation in a KDP crystal and subsequent post-compression of high-power laser pulses in relation to two cases: an initial pulse (1) with a duration of 20 fs with a central wavelength of 800 nm or 910 nm and (2) with a duration of 1000 fs or 300 fs with a central wavelength of 1054 nm. In the former case, the parameters correspond to the output parameters of most PW lasers [1–3] and almost all 100-PW projects [4–9]. The implementation of pulse post-compression at the second harmonic will in this case make it possible to modernize the corresponding facilities, significantly increasing the power and focal intensity of the output pulse. In the latter case, we are dealing with a fundamentally different approach to the architecture of high-power lasers with a duration of about 10 fs: the radiation of neodymium lasers is not used as a pump for parametric or Ti:sapphire amplifiers, but directly to generate fs pulses. In this case, the narrowband property of neodymium glass lasers is compensated by broadening the spectrum during post-compression, and frequency doubling (as will be shown below) for high-power neodymium lasers seems less difficult. Therefore, in relation to 100-PW-laser projects, for example, XCELS [9] or SEL-100PW [6–8], the second option represents an alternative concept that allows achieving the desired at-focus intensity in a much simpler, more compact and cheaper way.

2. Features of second harmonic generation under conditions of cubic nonlinearity and linear dispersion effects

The generation of the second harmonic of high-power femtosecond laser pulses in a nonlinear crystal is limited by a number of effects: the wavelength dependence of the refractive index of the crystal, self- and cross-interaction of waves, the development of small-scale self-focusing, self-steepening of the leading edge of the pulse due to the dependence of the group velocity on wave intensities, etc. We consider in more detail what these parasitic effects entail and discuss methods for suppressing them.

The wavelength dependence of the refractive index is a linear effect, which in the problem under consideration leads to the development of frequency modulation in pulses, as well as to a difference between group velocities of the first and second harmonic waves. Note that the phase velocities of the interacting waves coincide (or are at least close to each other) due to the fulfillment of the phase matching condition, which is necessary for the implementation of the second harmonic generation. The difference in group velocities between the first and second harmonic waves leads to the spread of pulses in time, which reduces the effective interaction length, reduces the conversion efficiency, and distorts the temporal distribution of intensity. The wavelength dependence of the group velocity leads to a broadening of the pulse in time, and the wider its spectrum, the greater the broadening. The influence of these effects is proportional to the thickness of the crystal. However, reducing the thickness of the crystal, with other parameters remaining unchanged, can lead to a decrease in conversion efficiency. Note that the central wavelength of the first harmonic pulse, equal to 1034 nm, is the dedicated wavelength for a KDP crystal, since observed in this case is the group synchronism — the equality of group velocities of interacting pulses. Pulses with a central wavelength of 1034 nm can be generated using active media doped with Yb^{+3} ions. In the general case, to optimize conversion efficiency, it is necessary to ensure the correct balance among the effects caused by quadratic and cubic nonlinearity, as well as the linear dispersion of the refractive index.

In the mode where the laser beam parameters are such that the contributions from the effects of quadratic and cubic nonlinearity are comparable, the frequency doubling differs from the classical case, i.e., when only quadratic nonlinearity of the medium is significant. Cubic nonlinearity leads to the accumulation of an additional, time-dependent (and space-dependent for nonuniform beams) phase. This phase, called the B-integral, is present in both the first and second harmonic waves and violates the phase matching condition. Contributions to the B-integral are made by both self- and cross-action of pulses in a medium with cubic nonlinearity. The value of the B-integral depends on the intensity at the input boundary of the crystal and the cubic nonlinearity tensor components, and is linearly proportional to the thickness of the crystal. It is significant that the contribution from the accumulated B-integral can be partially compensated by detuning the crystal from the phase matching direction. The optimal pulse interaction angle depends on the peak intensity at the input crystal boundary. To a first approximation, the dependence is linear [25]. In the model of interaction of plane monochromatic waves, complete compensation of nonlinear phase detuning is possible using a linear one [24]. However, in general, the optimal direction of wave propagation in a nonlinear crystal can be found by numerically solving a system of coupled equations that describe the frequency conversion or during experiments by adjusting the crystal in the plane critical to phase matching.

The manifestation of cubic nonlinearity during second harmonic generation is also responsible for the development of small-scale self-focusing [27, 28] in a high-power laser beam as it propagates in a nonlinear crystal. The essence of the phenomenon is the increase in the amplitudes of spatial harmonics falling within the amplification band. The boundaries of the amplification band are determined by the peak intensity and the cubic nonlinearity of the crystal. Fighting this parasitic effect involves minimizing the amplitudes lying in the amplification band, for example, due to self-filtering of the laser beam as it propagates in free space (vacuum). At a sufficiently long distance from the crystal, spatial harmonics escape from the region of the strong field of the laser beam both in time [12, 29] and in space [13, 30]. Note that, for laser radiation with a peak intensity of $\sim$ TW cm^{-2}, hazardous components propagate at an angle of tens of mrad to the direction of propagation of the laser beam [25]. For more details on methods to combat the development of small-scale self-focusing in intense femtosecond beams, see Ref. [13].

Another parasitic effect caused by cubic nonlinearity — self-steepening of the leading edge of the pulse — is associated with the intensity dependence of the group velocity. The higher-intensity part of the pulse in a medium with a positive cubic nonlinearity coefficient propagates more slowly than its wings. As a result, the pulse shape of both harmonics is distorted, which also negatively affects the efficiency of second harmonic generation.

2.1 Numerical modeling of second harmonic generation and post-compression of Nd:glass laser pulses

Pulses with energies of hundreds of joules or more with a duration of 1000 fs are routinely obtained in Nd:glass lasers using only one brand of glass. Shorter pulses with a duration of $\sim$ 150–300 fs can be obtained using a combination of silicate and phosphate glasses with different amplification spectra [31–34].

The spatial beam profile in high-power lasers is usually close to flat. The time profile is complex and determined by many factors, in particular, the consistency of the stretcher-compressor system, the amplifiers used, and the spectral characteristics of the optical elements of the laser system. Let the field envelope $A(t)$ at the input boundary of the crystal be of the form

$$A(t) = A_{10} \exp\left[-\left(\frac{x^2+y^2}{(D/2)^2}\right)^{2N}\right] F^{-1} \exp\left(-2\ln(2)\frac{\omega^2}{\Delta\omega^2}\right) + K \exp\left(-2\ln(2)\frac{\omega^2}{\Delta\omega^2}\right)\exp\left(\mathrm{i}\varphi(\omega)\right),$$

where A_{10} is the pulse amplitude, F^{-1} is the inverse Fourier transform operator, K is the coefficient that determines the level of temporal contrast, N is the degree of super-Gaussianity, and $\varphi(\omega)$ is the random spectral phase of the noise. Here, it is assumed that the spectral width of the noise component $\Delta\omega$ coincides with the spectral width of the signal. For modeling, we choose two values of pulse duration at the first harmonic: 1000 fs and 300 fs, with $K = 10^{-3}$, beam diameter $D = 20$ cm, and $N = 2$.

Apart from the pulse duration, the key parameter for both SHG and post-compression is intensity. In neodymium-glass lasers, it is usually limited by the optical strength of the diffraction gratings of the compressor, but, after the compressor, the intensity may be increased using an evacuated mirror telescope up to the breakdown threshold of the SHG crystal. We will consider the maximum permissible intensity to be 10 TW cm^{-2}, but 5 TW cm^{-2} is preferable and safer.

The results of numerical simulations for the specified parameter sets are presented in Figs 1–4. Solved numerically during the simulation was a system of quasi-optical coupled equations, which takes into account spatial effects: the angular drift of the extraordinary wave, diffraction, as well as dispersion effects, namely, pulse divergence in time, dispersion spreading, and manifestation of third-order dispersion. Also included were nonlinear effects: quadratic (SHG) and cubic (self- and cross-interaction of pulses) nonlinearity. An example of such equations (disregarding the third-order dispersion) is presented in Ref. [26]. Note that, for each crystal thickness, the angle of propagation of the laser beam to the optical axis was optimized. The optimization criterion was the highest energy conversion efficiency.

According to Fig. 1 for KDP crystals up to 0.5 mm in thickness and pulse durations of 300 fs or longer, the conversion efficiency is determined by the peak intensity, since dispersion effects are small. At the same time, for radiation with a duration of 20 fs (the central wavelength of the first harmonic is 910 nm), dispersion effects are important and result in a lowering of conversion efficiency.

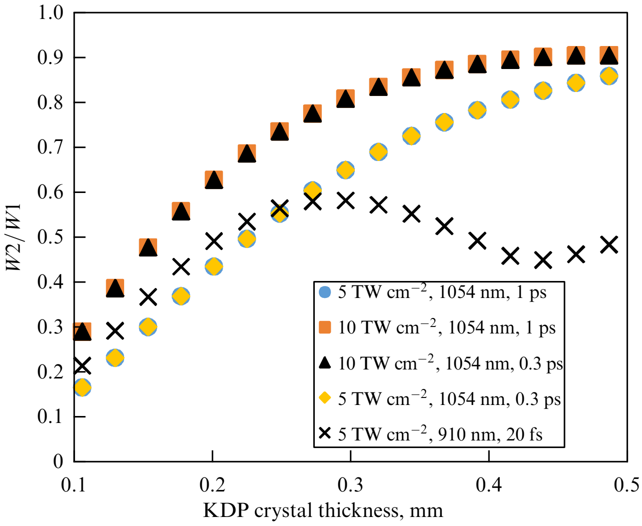

Figure 1. Efficiency of conversion to the second harmonic for pulses with a peak intensity of 5 and 10 TW cm^{-2} and a duration of 300 fs and 1000 fs with a central wavelength of 1054 nm, as well as for 20 fs, 5-TW-cm^{-2} pulses with a central wavelength of 910 nm.

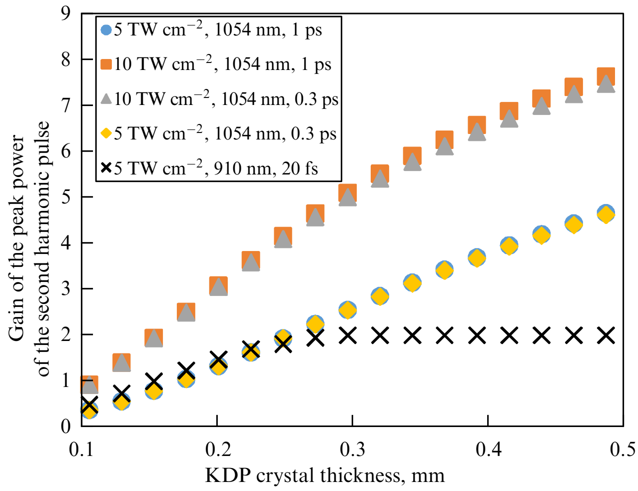

Figure 2. Dependences of the power gain factor F on the crystal thickness.

At the indicated intensity levels, the cubic nonlinearity of the KDP crystal leads to an enrichment of the spectrum of the second harmonic pulse, which can be used to further shorten the duration and increase the peak power with proper correction of the quadratic component of the spectrum phase. The dependence of the power gain factor $F = P_{out}/P_{in}$ (where $P_{in,\,out}$ is the input and output peak power) on the thickness of the KDP crystal is shown in Fig. 2. As a result of conversion to the second harmonic and correction of the spectrum phase, the peak power of the second harmonic pulse exceeds the peak power of the first harmonic pulse, even despite the lower pulse energy. With the parameters considered, the behavior of the curves is determined by the input peak intensity and is hardly dependent on the initial duration. All other parameters being equal, a higher peak intensity leads to a stronger manifestation of effects associated with cubic nonlinearity. In the problem under consideration, this is primarily a broadening of the spectrum that determines the duration of the second harmonic pulse after temporal compression.

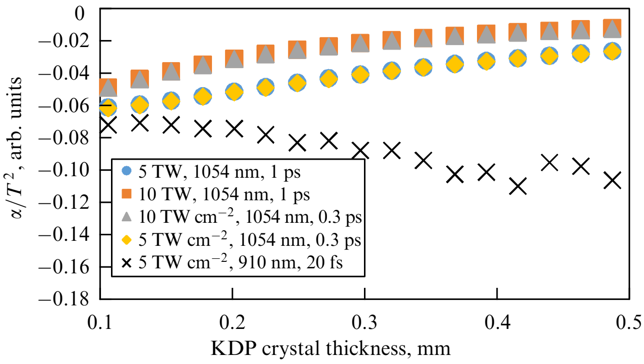

Figure 3. Optimal chirped mirror parameter α/T^2 required for correcting the phase of the spectrum of the second harmonic pulse.

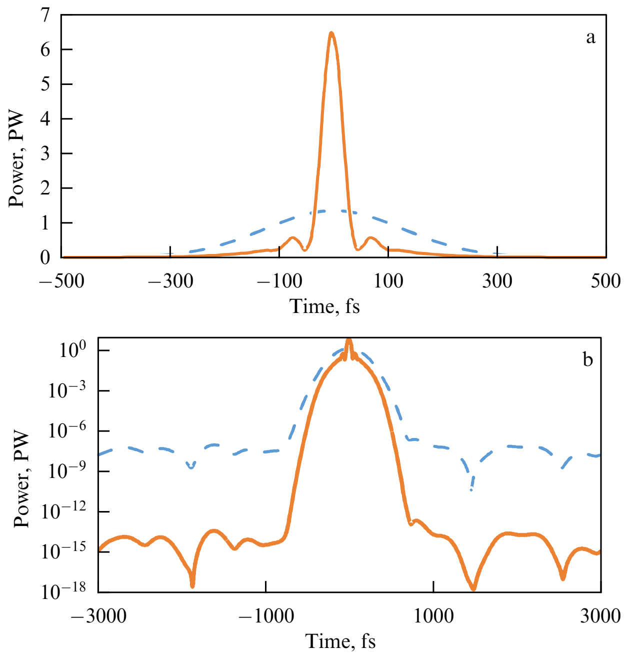

Figure 4. Normalized time profiles of first harmonic (dashed line) and second harmonic pulses after time compression (solid line) on linear (a) and logarithmic (b) scales.

The initial duration of the first harmonic pulse affects the value of the optimal quadratic dispersion of chirped mirrors α at which the maximum peak power is achieved. To correct the spectral phase of second harmonic pulses generated from longer pulses at the fundamental frequency, it is necessary to use chirped mirrors with a large absolute value of α. In this case, the combination of parameters α/T^2, where T is the duration of the spectrum-limited pulse of the first harmonic, is more universal, since it is determined only by the intensity level at the input to the selected nonlinear crystal. The optimal quadratic dispersion parameters α/T^2 are plotted in Fig. 3.

For initial 300-fs- and 1000-fs-long pulses, the values of $|\alpha|$ amount to tens of thousands of fs^2. It is important to note that the production of such mirrors with an aperture of 200 mm is a complicated technological task. Moreover, such mirrors must contain a large number of dielectric layers, which in turn reduces their radiation resistance.

2.2 Increase in temporal contrast during second harmonic generation of Nd:glass laser pulses

Next, we will consider an issue related to the increase in temporal contrast of second harmonic pulses after time compression. When the second harmonic is generated, the temporal contrast of the laser pulse at the doubled frequency is usually higher than that of the original pulse. The increase in temporal contrast is due to the nonlinearity of the conversion as well as the fact that noise frequencies lying outside the frequency doubling band are not converted into second harmonic radiation.

By way of example, in Fig. 4, we present the time profiles of the first and second harmonic pulses for one of the cases considered above (the first harmonic pulse: 5 TW cm^{-2}, 300 fs, 1054-nm central wavelength, 0.5-mm-thick KDP crystal).

The time contrast of the second harmonic pulse increases quadratically with respect to the time contrast of the original first harmonic pulse. The physical explanation of this fact is due to the second harmonic generation occurring in the unsaturated mode being quadratic in the field. This is precisely the situation that occurs in the wings of the pulse away from the main peak. The picture remains similar if the initial pulse of the first harmonic has a significantly shorter ($\sim$ 20 fs) duration.

3. Features of second harmonic generation by high-intensity ultrashort pulses

At present, in most laser facilities of petawatt power level, the radiation has a duration of tens of fs and a wavelength of 800 nm or 910 nm. To increase the energy per pulse, in the first case, use is made of laser amplification in sapphire crystals doped with titanium ions, and in the second, of noncollinear parametric amplification in DKDP crystals. The dispersion properties of the KDP crystal differ significantly for the indicated wavelengths. Specifically, the group divergence of the first and second harmonic pulses during the conversion of 800(910) nm to 400(455) nm is 77(36) fs mm^{-2}, i.e., the implementation of doubling at a wavelength of 910 nm is much simpler, regardless of the effect of cubic nonlinearity. In particular, for an initial pulse with a duration of 20 fs and a central wavelength of 800(910) nm, the use of KDP crystals thicker than 0.26(0.56) mm will lead to distortion of the temporal shape of laser radiation.

The production of large-aperture ($\sim$ 20 cm) crystals of so small a thickness is currently a technologically unsolved problem. The following solution was proposed in Ref. [9]. An $\sim$ 2–4-mm or thicker KDP crystal is glued onto an $\sim$ 1-mm-thick fused quartz plate using a transparent adhesive. Next, the surface of the KDP crystal is polished to the desired thickness of $\sim$ 0.25 mm. After SHG in the KDP crystal, a laser pulse at a doubled frequency hits the glued fused quartz plate, where it further broadens the spectrum. Further, reflection of the second harmonic pulse from the chirped mirrors makes it possible to correct the phase of the spectrum and compress the pulse in time. An example of numerical simulation of the operation of such a setup for converting laser radiation with a central wavelength of 910 nm, a peak intensity of 5 TV cm^{-2} and an initial duration of 20 fs is presented in Figs 5a, b.

One can see from Fig. 5 that a simultaneous increase in both the temporal contrast and the peak power of the second harmonic pulse is achieved. The increase in temporal contrast is slightly lower than the square of the initial value; however, the gain is significant. Moreover, this approach to second harmonic generation will potentially make it possible to experimentally generate laser pulses of petawatt power level in the visible range with a duration of several periods of the

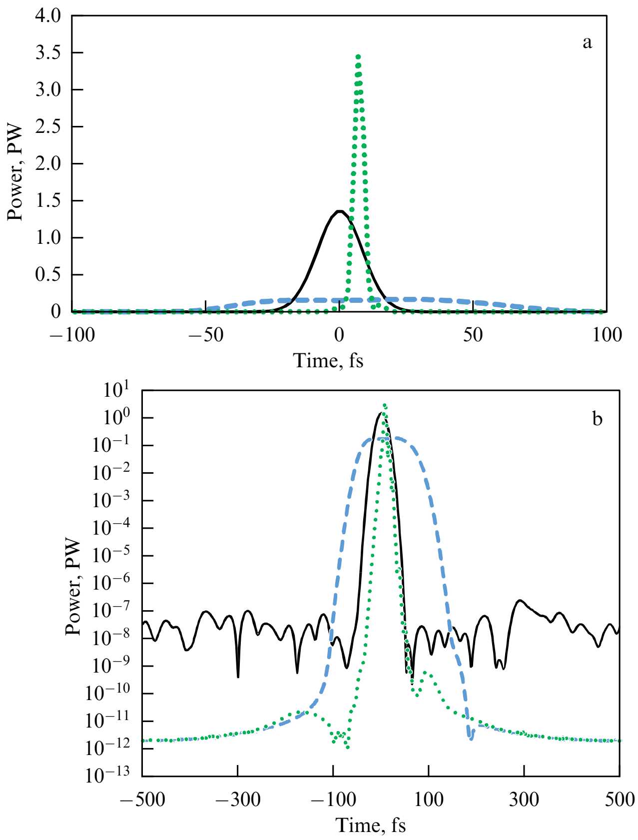

Figure 5. Time profiles of pulses: initial pulse (solid line), pulse at double frequency at the output of the KDP crystal (0.25 mm), and fused quartz (1 mm) (dotted line) structure, second harmonic pulse after temporary compression (dots). Linear (a) and logarithmic scale (b).

light field. In particular, in the example considered, the duration of the second harmonic pulse after time compression is $\sim$ 4 fs.

4. Conclusions

Our paper addresses the problem of using second harmonic generation with subsequent time compression of high-power laser pulses to improve their time characteristics. Frequency doubling can be implemented in a nonlinear KOR crystal at the output of an optical compressor. The process occurs under the influence of linear dispersion effects and effects associated with the manifestation of cubic nonlinearity. For pulses with a peak intensity of $\sim$TW cm^{-2}, which is characteristic of unfocused laser beams, the crystal must have a sub-millimeter thickness. To convert laser pulses of an ultrashort duration of $\sim$ 10–20 fs, it is also necessary to take into account linear dispersion effects. Minimizing their contribution requires an additional reduction in the crystal thickness to 0.25–0.5 mm. The additional use of chirped mirrors after the frequency doubling stage makes it possible to reduce the pulse duration to a few femtoseconds. Observed in this case is a significant increase in temporal contrast.

This concept can also be successfully applied to narrowband picosecond neodymium laser pulses. In this case, the radiation from neodymium lasers is not used to pump parametric amplifiers or Ti:sapphire amplifiers, but directly to generate fs pulses, which significantly improves the efficiency of the laser system. This approach is completely new and represents an alternative concept of 10–100-PW lasers. By virtue of this approach, it will be possible to achieve record intensity on a target in a much simpler, more compact and cheaper way.

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