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Highly efficient second-harmonic generation of intense femtosecond pulses with a significant effect of cubic nonlinearity

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Abstract. **A highly efficient (73%) second-harmonic generation of femtosecond pulses in a 1-mm-thick KDP crystal at a fundamental-harmonic peak intensity of 2 TW cm^{-2} has been demonstrated experimentally. In a 0.5-mm-thick KDP crystal, a 50% efficiency has been reached at a peak intensity of 3.5 TW cm^{-2}. We examine the key factors that limit the conversion efficiency and present numerical simulation results on further temporal compression of second-harmonic pulses.**

Keywords: *intense femtosecond pulses, second-harmonic generation, cubic nonlinearity.*

1. Introduction

Second-harmonic generation (SHG) finds wide application in petawatt laser systems [1–3]. Frequency-doubled laser radiation is used primarily to pump Ti-sapphire lasers and parametric amplifiers. Frequency conversion is also important for raising the peak intensity of focused light because it enables a twofold decrease in beam waist diameter. SHG can also be used to improve the temporal characteristics of ultra-high power femtosecond laser pulses. Of particular interest is the ability to reduce the pulse duration and increase the temporal pulse contrast [4]. Contrast enhancement is due to the fact that the second-harmonic generation efficiency is an essentially nonlinear function of light intensity. The second-harmonic pulse duration can also be reduced owing to the broadening of the spectrum (as a result of self- and cross-action effects) and subsequent phase correction using dispersion devices.

Optical frequency doubling is due to quadratic nonlinearity in crystals having no centre of inversion. An optical pulse propagating in a medium excites polarisation waves at frequencies that are multiples of the drive frequency. In classical nonlinear optics, efficient conversion requires phase matching between the generated second-harmonic wave and polarisation wave [5]. Phase matching occurs when the waves have equal phase velocities, which is possible at identical undisturbed refractive indices of the fundamental and second harmonics. Without phase matching, conversion efficiency is insignificant.

When petawatt radiation is frequency-doubled, an optical pulse generates not only waves with quadratic polarisation but also waves with cubic polarisation. Being in resonance with the fundamental and second harmonics, the latter waves give rise to an extra phase shift, known as the B integral [6, 7]. The high intensity induced refractive index modulation leads to a phase mismatch between the fundamental and second-harmonic waves [8–10]. Energy exchange can then be increased by correcting the light propagation angle in a nonlinear element by a value proportional to the incident intensity of the fundamental harmonic [4, 9, 11]. The resultant phase-velocity mismatch enables partial compensation for the cubic polarisation induced phase shift.

The effect of incident intensity on the optimal interaction angle was studied in Ref. [11]. The importance of taking into account the influence of cubic polarisation on the frequency doubling process was discussed in several reports [4, 8–10, 12, 13], but highly efficient (above 50%) SHG with a significant effect of cubic nonlinearity (at a B integral above unity) has not been reported to date.

This paper addresses the SHG of intense femtosecond pulses: the main factors that limit the conversion efficiency and experimental data on frequency conversion at peak intensities of up to 3.5 TW cm^{-2} in KDP nonlinear crystals. We present a comparative analysis of the experimental data and three-dimensional (3D) modelling results on frequency conversion. Numerical simulation is used to demonstrate that the pulse duration has a nonuniform distribution across the second-harmonic beam. We examine the possibility of second-harmonic phase correction with the aim of reducing the pulse duration and increasing the temporal pulse contrast for experimentally realisable laser beams.

2. Theoretical model for SHG in a very strong laser field

The frequency doubling of intense ultrashort pulses in the case of oo–e interaction can be described by the following system of differential equations [8, 14, 15]:

$$\frac{\partial A_1}{\partial z} + \frac{1}{u_1}\frac{\partial A_1}{\partial t} - \frac{\mathrm{i}k_2^{(1)}}{2}\frac{\partial^2 A_1}{\partial t^2} + \frac{\mathrm{i}}{2k_1}\Delta_\perp A_1$$

$$= -\mathrm{i}\beta A_2 A_1^* \exp(-\mathrm{i}\Delta kz) - \mathrm{i}\gamma_{11}|A_1|^2 A_1 - \mathrm{i}\gamma_{12}|A_2|^2 A_1,$$

$$\frac{\partial A_2}{\partial z} + \frac{1}{u_2}\frac{\partial A_2}{\partial t} - \frac{\mathrm{i}k_2^{(2)}}{2}\frac{\partial^2 A_2}{\partial t^2} + \frac{\mathrm{i}}{2k_2}\Delta_\perp A_2 + \rho\frac{\partial A_2}{\partial y}$$

$$= -\mathrm{i}\beta A_1^2 \exp(\mathrm{i}\Delta \boldsymbol{kz}) - \mathrm{i}\gamma_{21}|A_1|^2 A_2 - \mathrm{i}\gamma_{22}|A_2|^2 A_2, \quad (1)$$

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where A_1 and A_2 are the complex field amplitudes of the fundamental and second harmonics; z is a coordinate along the wave propagation direction; $\Delta k = k_2 - 2k_1$ is the wave vector mismatch; β and γ_{ij} $(i, j = 1, 2)$ are the second- and third-order nonlinear coupling coefficients [8]; ρ is the walk-off angle of the extraordinary wave; and $k_2^{(i)} = \partial^2 k_i/\partial^2\omega^2|_{\omega=\omega_i}$. The terms containing γ_{11} and γ_{22} represent the self-action of the fundamental and second harmonics, and the terms containing γ_{12} and γ_{21} represent cross-action effects. The system of equations (1) takes into account the difference in group velocity between fundamental (u_1) and second-harmonic (u_2) pulses, dispersion-induced pulse broadening, laser beam diffraction and the angular walk-off of the second-harmonic extraordinary wave. In the case of Gaussian pulses, the boundary conditions for system (1) have the form

$$A_1(\boldsymbol{r}_\perp, z = 0) = A_{10}(\boldsymbol{r}_\perp)\exp[-2\ln 2(t^2/T^2)],$$
$$A_2(\boldsymbol{r}_\perp, z = 0) = 0, \quad (2)$$

where T is the pulse duration and $A_{10}(\boldsymbol{r}_\perp)$ is the spatial distribution of the pulse amplitude.

Cubic nonlinearity results in detuning from the phase matching condition. The extra, nonlinear phase shift can be compensated for by changing the angle between the fundamental wave vector and the optic axis of the crystal by $\Delta\theta$. As shown previously [4], the angle detuning, $\Delta\theta$, that ensures optimal conversion for oo–e interaction in a plane monochromatic wave model is given by

$$\Delta\theta = \frac{\Delta n}{n_1^3(n_1^{-2} - n_o^{-2})}\sqrt{\frac{n_1^{-2} - n_o^{-2}}{n_e^{-2} - n_1^{-2}}}. \quad (3)$$

Here $\Delta n = \lambda_1 A_{10}^2(2\gamma_{11} + 2\gamma_{12} - \gamma_{21} - \gamma_{22})/(8\pi)$; n_1 is the refractive index at the fundamental frequency; and n_o and n_e are the principal refractive indices for the second harmonic. Expressions for the γ_{ij} $(i, j = 1, 2)$ of KDP crystals can be found elsewhere [4, 8]. For example, at an incident intensity of 3.5 TW cm^{-2} the angle detuning, $\Delta\theta$, needed for efficient SHG in a KDP crystal is –0.35°. The minus sign corresponds to a decrease in the angle between the light propagation direction and the optic axis of the crystal. It is important to note that taking into account the temporal and spatial beam structures reduces the compensation efficiency because the intensity and, hence, the increase in refractive index, Δn, are then functions of time and transverse coordinates.

An obvious implication of the above model is that optimal SHG conditions can be achieved in a quasi-static pulse interaction mode, when the length scale for the spatial separation between fundamental and second-harmonic pulses exceeds the characteristic conversion length. Moreover, in experimental studies of SHG in a strong laser field, one should ensure that self- and cross-action effects have an insignificant effect on the fundamental and second harmonic parameters.

3. Experimental results and discussion

To experimentally investigate frequency doubling in a strong laser field, we used the output beam from the front-end system of the PEARL petawatt femtosecond laser system [1]. Figure 1 shows a schematic diagram of a vacuum chamber for highly efficient SHG. Such experiments should be conducted in vacuum because the cubic nonlinearity of air has a strong effect at laser beam intensities of several terawatts per square centimetre. The influence of small-scale self-focusing on the SHG process was examined in detail by Ginzburg et al. [16]. The distances between optical elements were calculated using a self-filtration principle for harmonic disturbances of high-intensity laser beams [11]. Properly adjusted distances allowed us to avoid optical breakdown of the mirrors and KDP crystal even though the B integral exceeded 4.8 (for the second harmonic, $B = \gamma_{21}|A_{10}|^2L + \gamma_{22}|A_{10}|^2L$, where L is the thickness of the crystal).

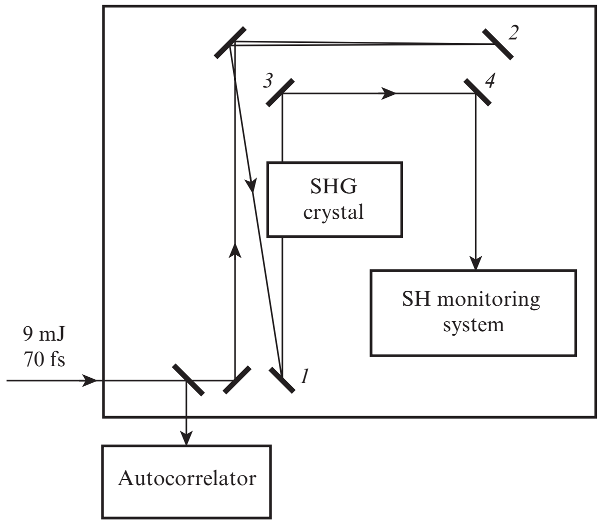

Figure 1. Block diagram of the pilot vacuum chamber for highly efficient SHG of intense femtosecond pulses: (*1*, *2*) spherical mirrors of a beam-contracting telescope; (*3*, *4*) wavelength-selective mirrors.

The fundamental-harmonic shaping/monitoring system comprised a mirror telescope (with a magnification factor $f_2/f_1 = 0.3$, where f_1 and f_2 are the focal lengths of the mirrors) for reducing the beam diameter, a femtosecond pulse width meter (second-order intensity autocorrelator), energy meter, CCD camera for measuring the beam profile and frequency spectrum analyser. The fundamental-pulse duration was measured before and after the experiments. The duration of each pulse is difficult to determine because a beam splitter (light transmission element) must be used, which considerably modifies beam parameters at intensities of several terawatts per square centimetre. The centre wavelength of the fundamental harmonic was monitored with a spectrometer and was ~910 nm in our experiments.

The frequency was doubled by KDP nonlinear elements 1 and 0.5 mm thick, cut at 42° to the crystal's optic axis. At the input of the KDP crystal, the pulse energy was 9 mJ and the full width at half intensity (for a Gaussian profile) was ~70 fs. In the problem under consideration, the phase-matching bandwidth at half maximum of the function $\mathrm{sinc}^2(\Delta kL/2)$ is 34 nm for the 1-mm-thick crystal. Note that the spectrum of a 36-fs transform-limited Gaussian pulse has the same width. The angular phase-matching bandwidth in the case under consideration is 9.36 mrad, which far exceeds the total laser beam divergence, 0.19 mrad, in our experiments. At these beam parameters, the group delay length is 1.9 mm, and the second-order nonlinear conversion length at an intensity of 3 TW cm^{-2} is 0.23 mm. The walk-off angle of the second-harmonic extraordinary wave is 28.8 mrad. Figure 2 illustrates the intensity distribution in the Fresnel zone in our experiments with the 1-mm-thick crystal. At a pulse energy of 9 mJ and pulse duration of 70 fs, the peak input intensity for this profile is 3 TW cm^{-2} and the average over the beam aperture is 1 TW cm^{-2}. The characteristic length scale for the spatial intensity distribution is $d = 4$ mm. For KDP crystals, the length scales on which the diffraction and angular walk-off of the second-harmonic extraordinary wave become significant are $k_1 d^2 = 166$ m and $d/(2\rho) = 69.4$ mm, respectively.

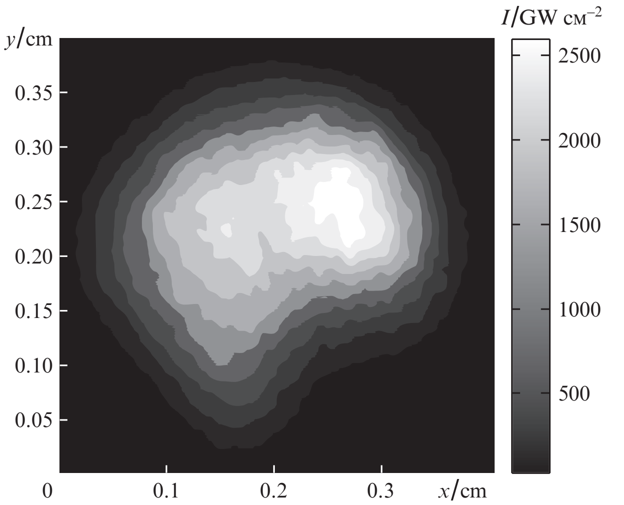

Figure 2. Fundamental-harmonic intensity distribution at the input of a KDP crystal.

The second-harmonic monitoring system comprised an energy meter, spectrum analyser and CCD camera in the Fresnel zone. The fundamental and second harmonics were separated by dielectric mirrors, which ensured a total fundamental-harmonic discrimination ratio of 10^{-4}. In energy measurements, we used calibrated pyroelectric detectors.

Figure 3a shows experimental and theoretical SHG efficiencies in the 1-mm-thick KDP crystal. In numerical simulation of the process, we used the basic equations (1), boundary conditions (2) and the fundamental-harmonic intensity distribution in Fig. 2. The angle detuning from the phase-matching angle was taken to be $\Delta\theta = 0.95$ mrad. The beam was assumed to have a Gaussian temporal profile.

As seen in Fig. 3a, increasing the fundamental-harmonic intensity increases the conversion efficiency to only a certain level. For the 1-mm-thick KDP crystal and the laser beam parameters under consideration, the optimal peak intensity is ~1.5 TW cm^{-2}. Further increase in energy density is accompanied by a reduction in conversion efficiency. The decrease in efficiency is caused by the cubic nonlinearity of the frequency doubling medium. As shown earlier [4, 11], angle detuning of a nonlinear element from the phase-matching angle allows one to raise the conversion efficiency for a top-hat laser beam profile. The optimal angle detuning depends on beam intensity. For laser beams similar to that used in our experiments (Fig. 2), with the angular walk-off of the second-harmonic extraordinary wave taken into account, detuning from the phase-matching angle increases the conversion efficiency only slightly (Fig. 4). The observed reduction in efficiency (Fig. 3a) at pulse energies above 6 mJ is due to the fact that high values of the B integral (which reaches 4.8 at this second-harmonic

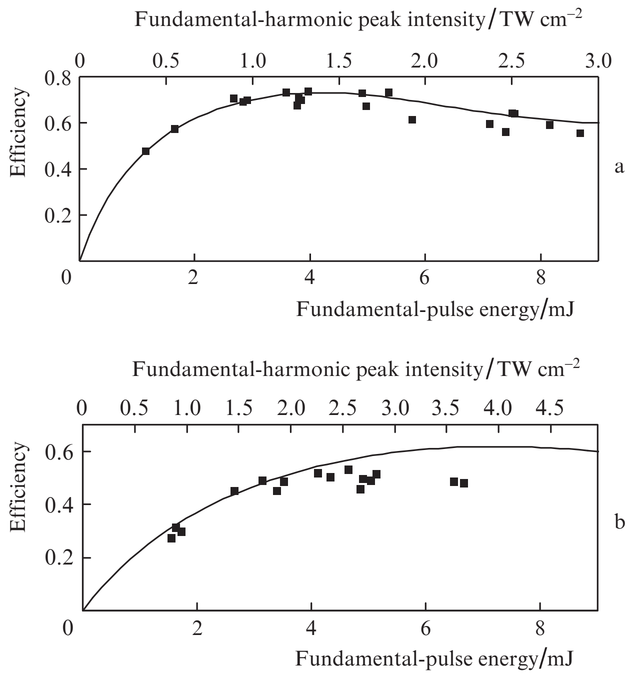

Figure 3. Experimental (filled data points) and theoretical (solid curves) SHG efficiencies as functions of fundamental-pulse energy and peak intensity at the input of KDP crystals (a) 1 and (b) 0.5 mm thick.

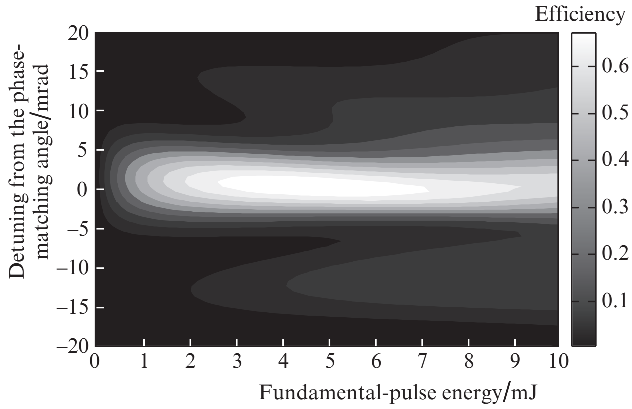

Figure 4. Conversion efficiency as a function of fundamental-pulse energy and detuning from the phase-matching angle for a 1-mm-thick KDP crystal.

pulse energy) lead to significant modulation of the spectrum. Typical fundamental- and second-harmonic spectra are presented in Fig. 5.

The effect of cubic nonlinearity can be reduced by lowering the input fundamental-harmonic intensity, which would increase the aperture of the crystal, and by utilising thinner nonlinear elements. Figure 3b shows the SHG efficiency in the 0.5-mm-thick KDP nonlinear element. In those experiments, the beam profile differed slightly from that in Fig. 2 in fill factor F (0.26 instead of 0.34), which can be represented as

$$F = \frac{\iint_{\Omega} I(x,y)\,\mathrm{d}x\,\mathrm{d}y}{\iint_{\Omega} \max[I(x,y)]\,\mathrm{d}x\,\mathrm{d}y},$$

where x and y are transverse coordinates and Ω stands for the region occupied by the beam. The reduction in the thickness

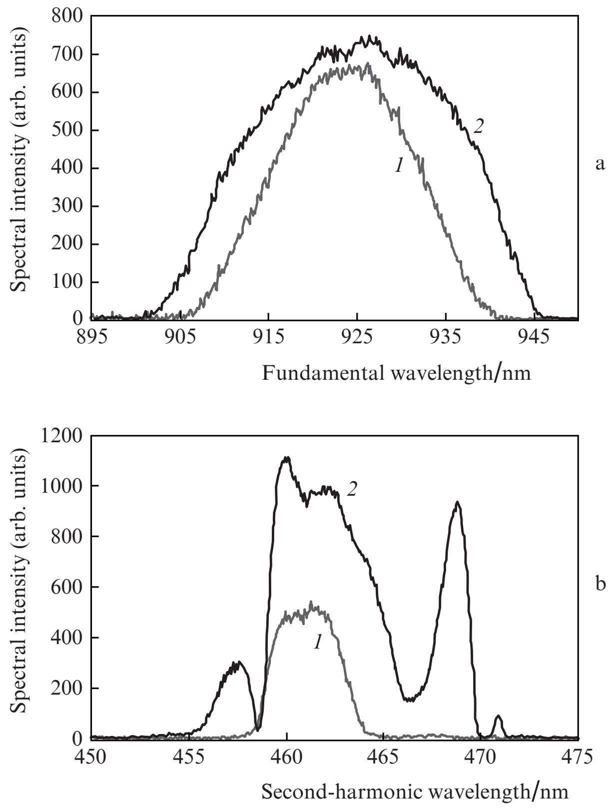

Figure 5. (a) Typical fundamental-harmonic spectra measured before the input of a nonlinear crystal and (b) the corresponding second-harmonic spectra in a 1-mm-thick KDP crystal. The fundamental-pulse energy at the input of the crystal is (*1*) 2.7 and (*2*) 4.7 mJ, and the *B* integral for the second harmonic is (*1*) 2.2 and (*2*) 3.7.

of the frequency doubling crystal led to a drop in efficiency by up to 50% but enabled the peak intensity at which cubic nonlinearity is insignificant to be raised to 3.5 TW cm^{-2}. In computer simulation, we used an experimentally determined beam profile and an angle detuning $\Delta\theta = -1$ mrad. The discrepancy between the simulation results and experimental data may be due to slight changes in beam shape during our experiments.

As seen in Fig. 3, the experimental data and simulation results are in reasonable agreement. Thus, we are led to conclude that the theoretical approach used to describe SHG with a significant effect of cubic polarisation is supported by experimental data for the beam parameters and nonlinear element thicknesses examined. At the same time, further raising the beam intensity requires that the fourth-order nonlinearity be taken into account. The use of shorter fundamental pulses makes it necessary to take into account transient crystal polarisation effects: the dependence of the group velocity on the intensity and the nonlinear susceptibility tensor dispersion [14].

4. Further temporal compression of second-harmonic pulses

Cubic nonlinearity of a frequency doubling medium gives rise to self- and cross-action of the interacting fundamental and second harmonics. This leads to spectral broadening (Fig. 5), and the pulses acquire phase modulation and are not transform-limited when emerging from the crystal. The phase modulation can be partially compensated through reflection from anomalous-dispersion (chirped) mirrors. Prism compressors cannot be used for further compression because they strongly modify intense pulses. Moreover, small-scale self-focusing may lead to optical breakdown in the glass. Experimental evidence of fundamental-pulse compression was reported by Mevel et al. [17], and a detailed theoretical analysis of this effect was presented by Akhmanov et al. [14].

Complete phase correction in experiments is a rather complicated problem, but even correction of the quadratic component enables a substantial decrease in pulse duration. Mathematically, this procedure can be represented by

$$A_{2c}(t) = \Phi[\exp(-i\alpha\omega^2/2)\Phi^{-1}(A_2(t,L))],$$

where $A_2(t,L)$ is the complex field amplitude of the fundamental harmonic at the output of the frequency doubling crystal; Φ and Φ^{-1} are the forward and inverse Fourier transformations; and α is the quadratic dispersion coefficient.

Figure 6 presents spatiotemporal dependences of the second-harmonic intensity at the output of a nonlinear element (frequency doubler) before and after correction of the quadratic component. The dependences were obtained by numerically solving the system of equations (1) subject to the boundary conditions (2), with the fundamental-beam profile shown in Fig. 2 (pulse duration, 70 fs; thickness of the KDP element, 1 mm). Phase correction enables a substantial decrease in second-harmonic pulse duration in the section through the average beam centre: from 54 fs at the output of the crystal to 35 fs. The α parameter was adjusted so as to minimise the pulse duration in this section. Note that the transverse second-harmonic pulse duration distribution at the output of a nonlinear element depends significantly on detuning from the phase-matching angle.

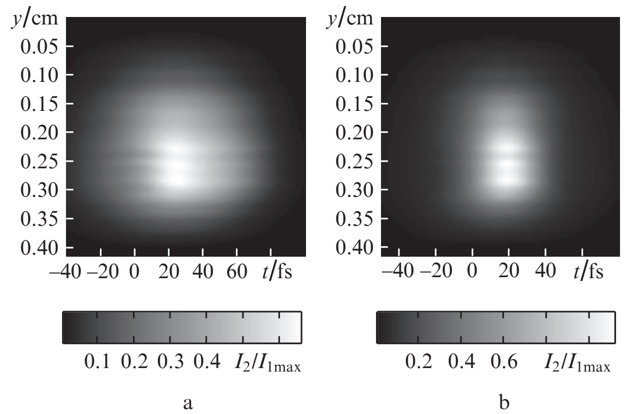

Figure 6. Spatiotemporal dependences of the second-harmonic intensity, I_2, normalised to the fundamental-harmonic peak intensity, I_{1max}, (a) before and (b) after quadratic phase correction: KDP crystal 1 mm in thickness; incident pulse energy, 5 mJ; pulse duration, 70 fs (the *B* integral is 4).

The spatial distribution of the fundamental-harmonic intensity at the input of a nonlinear crystal is typically nonuniform. Therefore, the accumulated *B* integral varies across the second-harmonic beam. As a consequence, correction of the quadratic component fails to ensure a pulse duration uniform across the beam. Numerical simulation of SHG with the beam profile shown in Fig. 2 indicates that, after further compression, the second-harmonic pulse duration may vary across the beam by a factor of 2. An increase in the fill factor of the fundamental beam would allow this effect to be reduced. Note that ultra-high power femtosecond laser pulses usually have a nearly top-hat beam profile.

Raising the fundamental-pulse energy increases the B integral, resulting in further spectral broadening. In the model under consideration, at a fundamental-pulse energy of 9 mJ and duration of 70 fs, second-harmonic pulses can be compressed to a 20-fs width at $\alpha = -202$ fs^2. It is worth emphasising here that, in this case, higher order terms in the Taylor expansion of the phase for the second harmonic remain uncompensated because the transform-limited pulse duration in a section through the average beam centre is 17 fs, i.e., 15% less than 20 fs.

5. Conclusions

Experimental data have been presented on the frequency doubling of pulses with a peak intensity of up to several terawatts per square centimetre. We have demonstrated a 73% energy conversion efficiency at a peak intensity of $\sim$2 TW cm^{-2} in a 1-mm-thick KDP crystal and a 50% efficiency at peak intensities of up to 3.5 TW cm^{-2} in a 0.5-mm-thick KDP crystal.

3D modelling of the SHG process allowed us to analyse the possibility of further pulse compression not only for model beams but also for an experimentally determined intensity profile. Theoretical analysis suggests that the second-harmonic pulse duration can be reduced from 70 to 20 fs.

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