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Shortening of the Femtosecond Pulse Duration during Second Harmonic Generation

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Abstract. The results of theoretical and experimental studies aimed at studying the possibility of reducing the second harmonic (SH) pulse duration during its generation under conditions of phase matching violation are presented in this paper. Generation is studied in a nonlinear KDP crystal pumped by transform-limited radiation pulse with a duration of 50 fs at a central wavelength of 950 nm. The efficiency of SH generation and the broadening of its spectrum as a function of the pump pulse energy and phase-matching angle detuning in different regions of the beam cross section are studied. The behavior dynamics of SH spectrum with a change in the phase-matching angle is shown, the optimal detuning angle corresponding to the maximum broadening of SH spectrum is determined. A nonlinear phase shift over SH beam cross section is noted, the sign of which depends on the sign of phase mismatch. The possibility of reducing femtosecond pulse duration by 1.5–1.7 times is shown.

Keywords: second harmonic generation, femtosecond pulse, phase matching angle, spectrum width, pulse duration.

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Introduction

Nowadays, to obtain high-power radiation pulses in visible spectrum range, high-power infrared (0.8–1 microns) laser systems are commonly used, the radiation pulses of which are converted into the second harmonic (SH) in nonlinear crystals. In Tomsk (IHCE SB RAS) together with the Moscow group (LPI RAS) an alternative way for obtaining such pulses in visible region based on THL-100 hybrid laser system is developed. This path is associated with the initial production of a femtosecond SH pulse in the visible region with low energies (1–5 mJ) and subsequent energy increase in gas amplifier based on XeF(C-A) molecules to the Joule level. THL-100 laser system operating on this principle is currently one of the most powerful systems in the visible spectrum range [1, 2]. The general interest in high-power radiation pulses in visible spectrum range is associated with higher energy of its quantum, which makes it possible to increase

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the interaction efficiency of laser beam with various materials and media. High-power pulses also help to develop new applications, which include the creation of X-ray laser in the water transparency window and generation of high-power terahertz radiation.

One of the ways to increase the power of THL-100 laser system is to reduce the pulse duration of output radiation while maintaining the radiation energy. In order to do this, it is required to increase the spectral width of SH radiation pulse in Ti:sapphire laser complex and amplify it in active medium on XeF(C-A) molecules, which have a wide amplification contour corresponding to transform-limited pulse duration of 10 fs [3]. Since the femtosecond complex operates at the amplification contour edge (950 nm) to match the SH wavelength with Ti:sapphire gas amplifier, it does not allow forming an SH pulse shorter than 50 fs (spectrum width 5–6 nm). To reduce transform-limited pulse duration of SH radiation, it is required to increase the width of its spectral contour by some artificial method. At present, two methods of spectrum broadening are known to reduce transform-limited pulse duration: due to high pump intensity of nonlinear crystal [4] and due to the conversion of chirped pump pulse in nonlinear crystal [5, 6].

It is known that during SH generation and other nonlinear optical transformations in media with quadratic nonlinearity, a nonlinear phase shift occurs when detuning from phase matching [7]. This shift is due to the cascade action of quadratic nonlinearity [8]. Such nonlinear process can lead to self-phase modulation, spectrum broadening, and shortening of SH pulse duration.

In this paper, the results of theoretical and experimental investigations aimed at studying the possibility of shortening the SH radiation pulse duration when pumping a nonlinear KDP crystal by femtosecond transform-limited pulses under conditions of phase-matching violation.

1. The equipment and methods

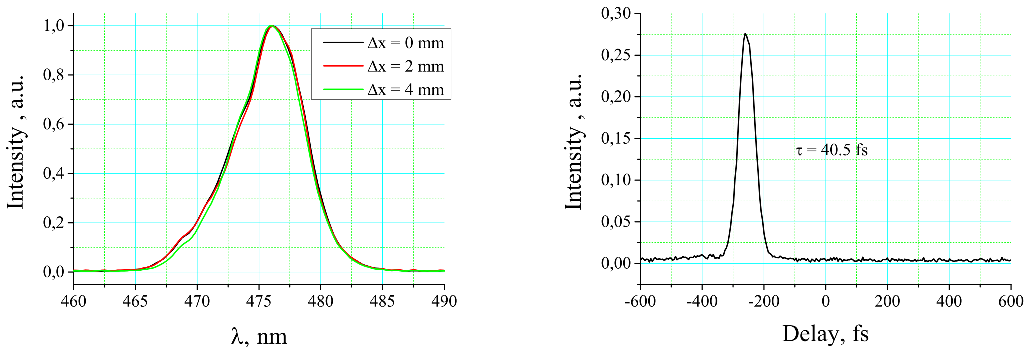
A femtosecond Ti:sapphire laser complex operating at the edge of amplification contour (central wavelength of 950 nm) was used in experiments. This complex is part of THL-100 multiterawatt laser system and consists of master oscillator, stretcher, regenerative and two multipass amplifiers, and compressor on diffraction gratings. The output pulse at fundamental harmonic has 50 fs duration, Gaussian profile with diameter of 12–14 mm in intensity decay by a factor of e^2, and energy up to 10 mJ. To obtain SH, a transform-limited pulse of fundamental harmonic was directed to nonlinear KDP crystal 2 mm thick with phase-matching of the first type. In the experiments, phase-matching angle was changed by tilting the crystal forward or backward in the direction of pump beam propagation. The spectrum was recorded in different regions of the SH beam cross section using diaphragm with diameter of 1 mm. The laser radiation energy was recorded with Gentec maestro energy meter. The emission spectra were recorded with spectrometers ASP150C (Avesta project) and Ocean Optics HR4000 (200–1100 nm, spectral resolution 0.7 nm).

Numerical simulation of the dependence of SH parameters in case of direction deviation of pump radiation propagation at the fundamental frequency from phase-matching angle was performed using the technique described in [8].

2. Results and discussion

In studying the broadening of SH beam spectrum in different regions of cross section, the spectrum as a function of phase-matching angle change and pump radiation intensity was measured. At the maximum efficiency of radiation conversion into the second harmonic (pump energy 10 mJ), the spectral width was about 6 nm (Fig. 1a). In this case SH beam had the following parameters: Gaussian profile, diameter of 12 mm with respect to the intensity decay by factor of e^2, energy of 2.2 mJ (conversion efficiency 22 %), and pulse duration (FWHM) of

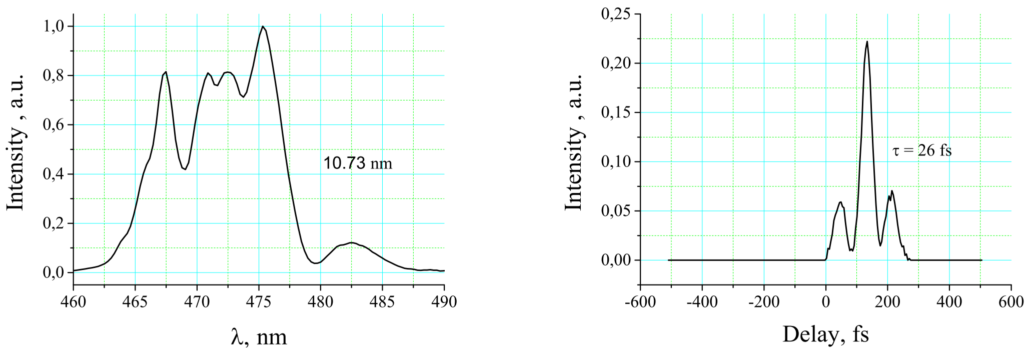
40.5 fs (Fig. 1b). The SH emission spectrum in different sections of cross section in vertical and horizontal planes remained virtually unchanged (Fig. 1a).



(a) (b)

Fig. 1. Spectrum over the beam cross section at different distances (0, 2, and 4 mm) from the optical axis (a) and autocorrelation function (b) of SH when pumping of nonlinear crystal under phase-matching conditions

When the crystal was tilted against the direction of pump beam propagation in the vertical plane, the SH spectrum was observed to broaden toward shorter wavelengths, and at tilt angle of $\sim 0.3°$, the spectrum width increased almost by a factor of 2 (Fig. 2a). In this case, the decrease of conversion efficiency by a factor of 3.6 was observed (SH energy of 0.6 mJ). At the maximum width of the spectrum, amplitude modulation of the SH radiation intensity was observed. The SH pulse duration (FWHM) was reduced to 26 fs (Fig. 2b).



(a) (b)

Fig. 2. Spectral contour (a) and autocorrelation function (b) of SH radiation pulse when the phase-matching conditions are violated (crystal tilt by 0.3°)

Theoretically, the dependence of generation efficiency and pulses duration of SH radiation was calculated when the phase-matching conditions were violated (Fig. 3). Under the phase matching conditions, the maximum conversion efficiency ($\sim 60\%$) was observed at pump pulse duration of 50 fs and pump energy of 10 mJ. The minimum pulse duration of SH radiation was about 26 fs and one was achieved at deviation angles of direction propagation of pump radiation from the matching direction by $\sim 0.3°$, both on the beam axis (blue curve) and for the entire beam (red curve). In this case, the conversion efficiency decreased by almost 10 times.

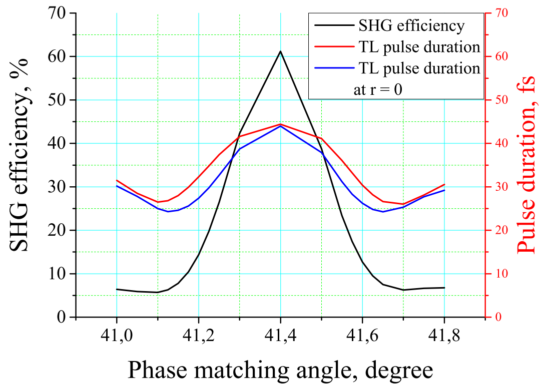


Fig. 3. Dependences of the SH generation efficiency (black curve) and the duration of a transform limited (TL) pulse (red and blue curves) upon deviation from the phase-matching angle

It is well known that during the generation of second harmonic, when detuned from phase matching, the nonlinear phase shift occurs [7], the sign of which depends on the phase mismatch sign. This shift is due to the cascade action of quadratic nonlinearity and one can be described by effective third-order nonlinearity [8]. Accordingly, such nonlinear process can be cause of self-phase modulation, which leads to broadening of second-harmonic pulse spectrum and under certain conditions to reduction in its duration. Also, the nonlinear phase shift can lead to reverse pumping of converted radiation into pump radiation and, thereby, limit the conversion efficiency. It should be noted that since the nonlinear phase shift is also determined by pump radiation intensity, the spectrum broadening and pulse duration decrease of converted radiation over the beam cross section will be inhomogeneous [9].

The formula for calculating of nonlinear cascade phase is presented in [8], it is correct for conversion efficiency up to 30 %:

$$n_{2,casc} = \frac{\pi \Delta k d_{eff}^2}{\epsilon_0 c \lambda n^3 Q^2}[1 - \frac{sin(QL)}{2QL}], \tag{1}$$

where $Q = \sqrt{2\sigma^2 E^2 + \Delta k^2/4}$, L is the crystal length, $\sigma = 2\pi d_{eff}/\lambda n$, Δk is the wave detuning, d_{eff} is the effective quadratic nonlinearity coefficient, λ is the central wavelength, E is the electric field.

It follows from the presented formula that compared with Kerr cubic nonlinearity the effective coefficient of cascade quadratic nonlinearity has a sharp dependence on wavelength in accordance with dependence of wave detuning. Therefore, the broadening of second harmonic spectrum is asymmetric when detuned in different directions from the phase matching angle. Moreover, as the conversion efficiency ($\sigma E \sim \Delta k$) increases intensity dependence appears i.e. the nonlinearity ceases to be cubic and at $\sigma E \gg \Delta k$, the magnitude of nonlinearity tends to zero with increasing intensity. Those, under phase matching when the wave detuning value Δk is close to zero the broadening of second harmonic spectrum does not occur, which is what we see in the experiment.

Fig. 4 shows the spectral curves of second-harmonic radiation at different crystal tilt angles. When SH is generated with maximum efficiency under phase-matching conditions, the spectral profile is uniform over the beam cross section (Fig. 4a). In this case, the duration of spectrally limited SH pulse is 44.4 fs at high conversion efficiency. All this agrees well with the experimental data (40.5 fs). Fig. 4 (b, c) shows the shift and modulation of spectrum when the crystal is tilted in different directions by the same amount (0.3°). It can be seen that the broadening of spectral contour is asymmetric. At the angle $\theta = 41.7°$ (Fig. 4, b), the radiation spectrum on axis and total spectrum broaden to the long and short wavelength regions respectively. When tilted to the other side (Fig. 4c) from the optimum by the same value (0.3°), the broadening of entire beam and central part occurs mainly in the short-wavelength region. In this case, the spectrum shape differs significantly over the cross section of second harmonic beam and depends on intensity of fundamental radiation at the considered point (Fig. 4d). The farther from beam center the greater spectrum broadening is observed. The conversion efficiency drops to 5–6 % with broadening. The duration of SH radiation pulse at spectrum maximum broadening decreased from 44.4 to 26 fs which is also in good agreement with the experimental results.

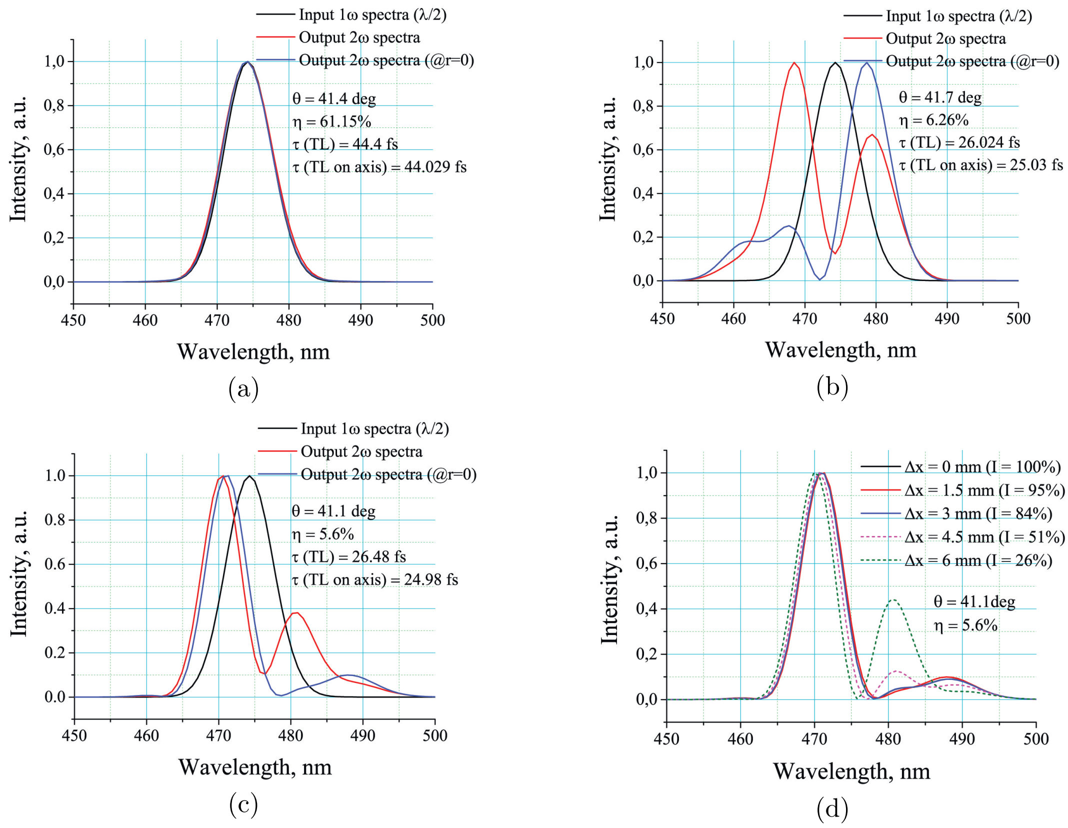


Fig. 4. Spectral profiles of second-harmonic radiation at different crystal tilt angles (a, b, c) and at different distances from the optical axis (d). For (a, b, c) the black line is the spectrum for $0.5\lambda_{pump}$, the red line is for the entire beam, and the blue line is on the beam axis

Conclusion

Thus, the experimental and theoretical studies have shown the possibility of radiation spectral contour broadening and reducing pulse duration of the second harmonic at central wavelength of 475 nm in nonlinear KDP crystal 2 mm thick due to the violation of phase matching. Sufficiently good agreement is obtained between experimental and theoretical results in terms of deviation from matching angle, pulse duration, and spectrum width. The pulse duration of SH radiation decreased by factor of 1.7 in calculations and by factor of 1.5 in the experiment. The optimal conditions for reducing the spectrally limited pulse duration of SH were fulfilled by changing the incidence angle of pump beam on nonlinear crystal from the phase-matching angle by $\sim 0.3°$. It is shown that due to the nonlinearity of spectrum broadening due to phase shift the decrease in pulse duration of converted radiation over the beam cross section is inhomogeneous. It can be noted that there is a difference between the theoretical and experimental results in terms of SH generation efficiency.

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Сокращение длительности фемтосекундного импульса при генерации второй гармоники

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Аннотация. В работе приводятся результаты теоретических и экспериментальных исследований, направленных на изучение возможности сокращения длительности импульса второй гармоники (ВГ) при её генерации в условиях нарушения фазового синхронизма. Генерация исследуется в нелинейном кристалле КДП при его накачке спектрально ограниченным импульсом излучения с длительностью 50 фс на центральной длине волны 950 нм. Исследуется эффективность генерации ВГ и уширение её спектра в зависимости от энергии импульса накачки и расстройки угла синхронизма в различных областях сечения пучка. Показывается динамика поведения спектра ВГ при изменении угла синхронизма, определяется оптимальная величина угла расстройки, соответствующая максимальному уширению спектра ВГ, отмечается нелинейный сдвиг фазы по сечению пучка ВГ, знак которого зависит от знака фазового рассогласования, и показывается возможность сокращения длительности фемтосекундного импульса в 1.5–1.7 раза.

Ключевые слова: генерация второй гармоники, фемтосекундный импульс, угол синхронизма, ширина спектра, длительность импульса.