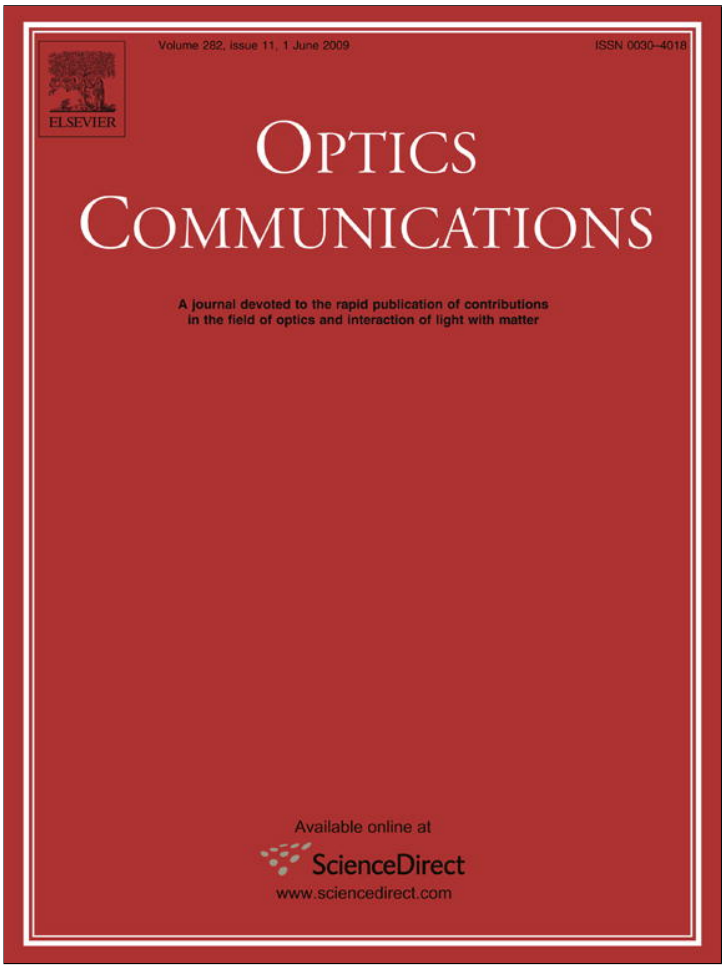

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Tunable femtosecond frequency doubling in random domain structure of strontium tetraborate

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ABSTRACT

Efficient tunable femtosecond phase-matched noncollinear second-harmonic generation in randomized nonlinear photonic crystal of strontium tetraborate is obtained. Spatial spectrum of nonlinear photonic structure is not flat but enables tuning of the harmonic frequency in the range from 355 to 460 nm. The narrowing of the bandwidth of second harmonic is found to be of order of 10–20%.

1. Introduction

Periodically poled nonlinear crystals (PPNC) are widely used for achieving quasi-phase matching (QPM) in those cases when angular phase matching is absent or suffers from limitations [1]. QPM requires the presence of domains with alternating oppositely poled static polarization, which appear as periodic spatial modulation of the sign of the second-order nonlinear susceptibility. Ferroelectric crystals potassium titanyl phosphate (KTP) and $LiNbO_3$ are most suitable for implementation of PPNC. However, their transparency in the ultraviolet (UV) is limited to 350–330 nm. For nonlinear conversion into UV, borates are commonly used; however, they have lower nonlinear susceptibility and suffer from some limitations, too. For instance, phase-matched direct second-harmonic generation in BBO can be obtained at wavelengths down to 210 nm with the effective nonlinearity approaching to zero, while the transparency limit is about 190 nm for this crystal. Tunable efficient femtosecond sum frequency generation of vacuum ultraviolet (VUV) was achieved in another borate crystal, LBO [2]. The only disadvantage of this scheme is the complication of laser installation that, for the sake of achieving angular phase matching, must generate two wavelengths and requires powerful femtosecond laser source. This complication can be overcome using ferroelectric domain structured crystals due to QPM. However, to avoid influence of narrow bandwidth typical for QPM, domain structure must be randomized; the reciprocal superlattice vector (RSV) spectrum of properly randomized structure may be flat in any required range of values, or present any other desired dependence. Recently a lot of studies on random two-dimensional domain structures in strontium barium niobate (SBN) were done. Conical emission of the second harmonic in nanosecond regime was investigated in SBN [3]. Both conical and planar emissions of femtosecond pulses were studied in [4], and spatially integrated conversion efficiency obtained for SBN, using low power femtosecond oscillator as the pump source, was 0.38%. Tuning of the SH frequency from 430 to 680 nm in SBN was demonstrated in [5]. Noncollinear SHG was employed for the diagnostics of microstructure of SBN crystal in [6]. Using periodic circular domain structure of lithium tantalate, conical SH emission was obtained in [7]. In many cases conical emission can be considered as disadvantage, if unidirectional beam of SH is desirable. In such cases 1D structures must be used. Another possible disadvantage of SBN is the transparency limit lying at 400 nm. Noncollinear SHG of femtosecond pulses was also investigated in one-dimensional periodically poled lithium niobate [8]; however, efficiency was found to be rather low.

Recently, domain structures consisting of alternate oppositely poled domains were found in strontium tetraborate (SBO) [9]. Transparency of this crystal in the VUV allows nonlinear generation of radiation with wavelengths as short as 125 nm [10]. Noncollinear second-harmonic generation (nonlinear diffraction) in SBO was studied in nanosecond pulse duration range [11]. Enhancement of collinear second-harmonic generation due to random QPM [12,13] in SBO was investigated in [14]. SBO belongs to *mm2* symmetry point group, and its domain structure is similar to that of KTP that belongs to the same symmetry group. Notation of crystallographic axes of SBO used below corresponds to $Pnm2_1$ space group, i.e. $a = 4.4145$ Å, $b = 10.6827$ Å, $c = 4.2234$ Å.

Presently SBO is not considered to be a ferroelectric, and as-grown structures that form at temperature of growth (in the vicinity of 1000 °C) are not well controllable. These domain structures represent one-dimensional nonlinear photonic crystal with domain walls perpendicular to *a* axis of the crystal and with homogeneous distribution of linear refraction index over the samples. The structures are well ordered in *b* and *c* directions at distances up to 1 cm, thickness of domains being highly randomized and lying in

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the range from tens to tenths of micrometer. The description of domain structures statistics typical for SBO can be found in [14]. Properties of domain structures relevant to nonlinear conversion processes can also be directly established by the analysis of angular dependence of nonlinearly diffracted energy. Such study for SBO at fixed fundamental wavelength (1064 nm) [11] allows to estimate the behavior of RSV spectrum and to predict the spectral range of fundamental wave where efficient nonlinear diffraction can be observed. This analysis predicts that typical domain structures grown up to date will enable efficient nonlinear diffraction at fundamental wavelengths down to 450–500 nm. At shorter wavelengths efficiency of nonlinear diffraction does not drop to zero but becomes less efficient. The maximal nonlinear coefficient in SBO is d_{ccc} that dictates the preferable orientation of fundamental wave polarization. Hence, preferable direction of fundamental radiation for nonlinear diffraction process must coincide with *b* axis. RSV vector direction coincides with *a* axis, and nonlinearly diffracted radiation propagates as beams lying in *ab* plane of the crystal.

In this letter we report the observation of femtosecond second-harmonic generation (SHG) in nonlinear photonic SBO crystal structure with random domains. In our experiments we used a femtosecond Ti:sapphire oscillator Spectra Physics Tsunami pumped by 5 W MilleniaPro. Pulse width was 40–100 fs full width at half maximum (FWHM), with average power up to 1 W, pulse energies up to 12.5 nJ, repetition rate of 80 MHz, and tunability in the range of 710–930 nm with the maximum of average power at 800 nm. The beam from the laser is focused inside the $6 \times 6.5 \times 5$ mm^3 SBO crystal by a 10 cm focal length lens, resulting in a focal spot of 100 m and peak intensity up to 4 GW/cm^2. The spectral properties of the fundamental and generated second harmonic are measured by Ocean Optics HR4000 spectrometer with 0.75 nm resolution. Fundamental radiation propagated along *b* crystallographic axis, and its polarization coincided the *c* axis (Fig. 1). Nonlinear photonic crystal structure in the sample under study was investigated in [11] and is shown to be highly randomized. It does not occupy all the volume of the crystal sample, but has the thickness of 2 mm in the direction of *a* axis. Due to high degree of randomization, the RSV spectrum of the structure may vary over its volume. Fundamental beam waist is sufficiently smaller than the thickness of nonlinear photonic crystal. For these reasons, the translation of the fundamental beam along *a* axis was employed in the course of measurements, together with small rotation tuning around *c* axis.

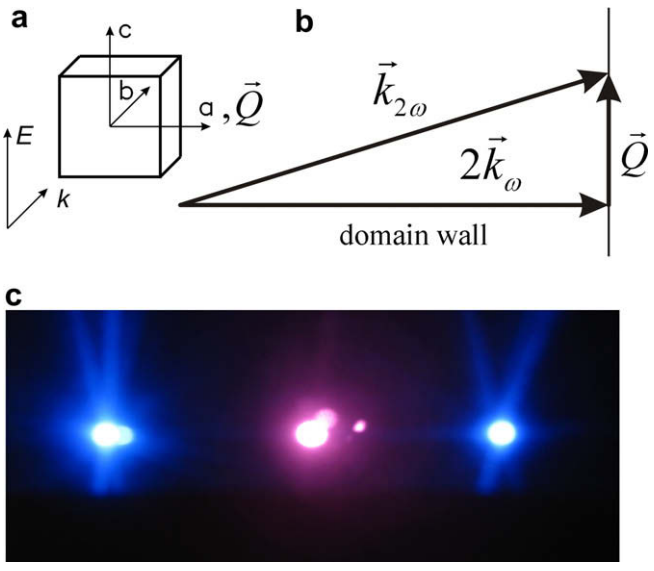

Fig. 1. (a) Geometry of fundamental wave propagation; (b) phase matching diagram; (c) nonlinear diffraction pattern of second harmonic radiation from SBO. The fundamental wavelength is 800 nm, with average power 1 W.

The observed pattern of nonlinear diffraction under fundamental beam propagation along *b* axis fairly agrees with expected one (see Fig. 1). Polarization of generated SH is found to coincide with both polarization of fundamental and c axis of the crystal. For zero incidence angle of fundamental beam external nonlinear diffraction angle θ_2 equals to 16.6°. This value is in fair agreement with the value of 16.9° calculated according to formula [11]

$$\theta_2 = arc\sin\sqrt{n_2^2 - n_1^2 + \sin^2\theta_1} - \theta_1 \quad (1)$$

where θ_1 is the external angle of fundamental wave incidence on the sample facet perpendicular to the domain walls, and $n_{1,2}$ are refraction indices for fundamental and harmonic waves, respectively.

Under the central spot produced by fundamental beam the spot of phase-mismatched collinear generation is present, with intensity much smaller than those of nonlinearly diffracted beams. Polarization of generated radiation coincides with that of fundamental, i.e. d_{ccc} component of second-order susceptibility is employed. Removing the crystal from the beam and introducing it back, we did not found any observable changes in the shape of the fundamental beam. So we conclude that thermal focusing is absent for the regime used in our experiment, in contrast to SBN crystal. Maximum average power of second harmonic detected in our experiment is 5.6 mW per both beams at 400 nm, with the efficiency 0.57%, the value being higher than that obtained for SBN, despite nonlinear susceptibility of the latter is much higher. This is due to RSV spectrum of the sample under study, since peak intensity in SBN experiment was higher due to self-focusing. Quality of the second harmonic beam in the near field is rather good.

Fig. 2a (squares) presents the tuning curve of the sample under study, i.e. the dependence of second harmonic power on the central wavelength of the laser. The fundamental beam position was optimized to maximize harmonic output in every measurement point. Additionally, rotation of the sample by small angles of order

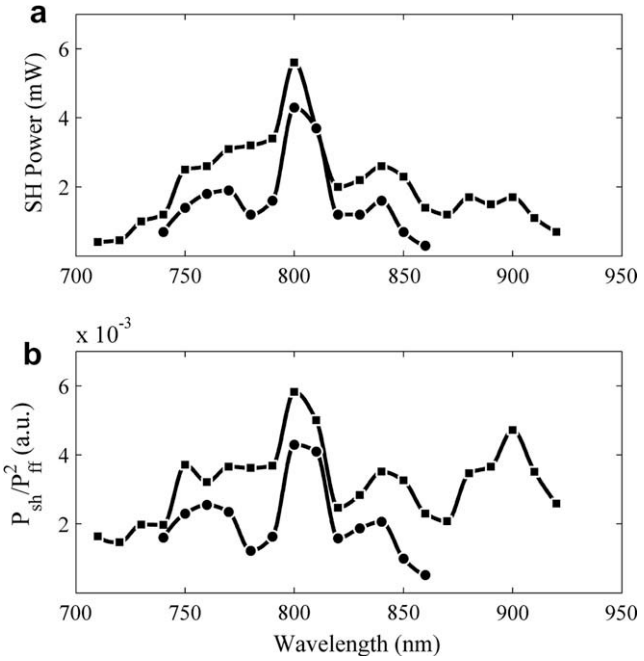

Fig. 2. (a) Spectral dependence of second harmonic power by tuning the femtosecond Ti:sapphire oscillator; (b) RSV spectrum of SBO nonlinear photonic crystal; squares – with translational tuning of second harmonic, circles – for fixed position of the beam.

of several degrees was used for optimization together with translation, in order to find the best part of RSV spectrum. However, the effect of rotation was proven to be small. The tuning curve of the laser itself is not removed from the dependence in Fig. 2a. As one can see, tuning curve of the harmonic generally resembles that of the laser. However, local peaks and minima of this dependence evidence that RSV spectrum is not smooth in the range of values involved in the nonlinear diffraction in the given wavelength region. Fig. 2a (circles) presents the tuning curve for a stable position of the fundamental beam against the nonlinear photonic structure, initially tuned to the maximum SHG at 800 nm. It demonstrates that selected part of the crystal contains most of the RSV spectrum necessary for efficient conversion, with exception for certain spectral regions where RSV spectrum is more favorable in other parts of domain structure. Fig. 2b presents the second harmonic power normalized to the square of fundamental power. This dependence clearly represents the RSV spectrum of nonlinear photonic crystal averaged over the bandwidth of the laser. Fig. 2b admits that for SBO this spectrum is wide but not flat. Especially large variation is observed at RSV values responsible for nonlinear diffraction in the longer wavelength part of the tuning curve.

Fig. 3 presents spectrum of fundamental radiation and that of second harmonic. For convenience of comparison the latter is reduced to the wavelength scale of fundamental. Spectrum of fundamental is taken at 840 mW average power in the beam that passed SBO crystal. Contrary to SBN, fundamental spectrum bears no signs of phase self-modulation and broadening caused by it. Spectrum of harmonic is evidently narrower than that of fundamental, however, this narrowing is of order of 10–20% for different wavelengths. When fundamental wave spectrum falls on a slope of RSV spectrum, also slight shift of the maximum of harmonic spectrum is observed. Narrowing of harmonic spectrum is well understandable and is due to non-flat RSV spectrum. Note that the narrowing extent is small, that evidences that real RSV spectrum, being not averaged over laser bandwidth, does not strongly differ from averaged one in Fig. 2c. Relative smoothness of harmonic spectrum with respect to that of fundamental is caused by worse energy resolution of spectrometer in shorter wavelength region.

During the tuning curves measurements, the duration of laser pulses at the entrance of the nonlinear medium could not be sustained absolutely constant and might influence the results. To verify the extent of this influence, we examined dependence of SHG coupling coefficient on the fundamental pulse duration. The latter was varied by changing the spectrum of Tsunami laser radiation, since design of this laser enables generation of transform limited pulses, duration of them being monitored via calibration curves supplied by the producer. As can be seen from Fig. 4 taken at 790 and 850 nm fundamental wavelengths, with account for variation of average power, this dependence is very weak in the range from 40 to 90 fs. As expected both from theory [15] and known experiments for collinear femtosecond harmonic generation, this dependence must follow approximately inverse proportional law with the variation of duration pulse. Explanation of observed experimental dependence, to our opinion, can be found in non-collinear character of SHG, however, this possibility needs additional extensive theoretical studies. However, results presented in Fig. 4 allow us to conclude that unintended variation of pulse duration during wavelength tuning does not seriously affect the results plotted in Fig. 2. Duration of the second harmonic pulses in the conversion process is expected to increase, both due to narrowing of spectrum and, to a greater extent, due to group velocity dispersion. Typical value of the latter in SBO equals to $2.8 \cdot 10^{11}$ cm/s, leading to harmonic pulse duration calculated for non-stationary regime [15] of order of 2 ps for the crystal length used in our experiment. However, this value can be decreased when shorter crystals are used, with a compromise between shortening of pulse duration and lower efficiency. At 1 mm crystal one may expect 350 fs pulses of second harmonic.

Results obtained in our study can be transferred into shorter-wavelength region, leading to creation of compact tunable source of femtosecond radiation in the region of wavelengths 200–175 nm. Two conditions must be satisfied for this, namely, (1) the second harmonic of Ti:sapphire oscillator introduced into SBO crystal must be of order of 1 W, and (2) characteristic domain sizes in nonlinear photonic crystal structure must be scaled down by value of order of 2. The first condition seems to be realistic at the present state of the art of laser technology, while second one needs further studies.

In conclusion, an efficient tunable SHG of femtosecond pulses from Ti:sapphire oscillator in randomized nonlinear photonic crystal structure of strontium tetraborate is demonstrated. Tuning range of second harmonic is from 355 to 460 nm. RSV spectrum of nonlinear photonic structure is not completely flat but enables second-harmonic generation with minimal narrowing of spectrum.

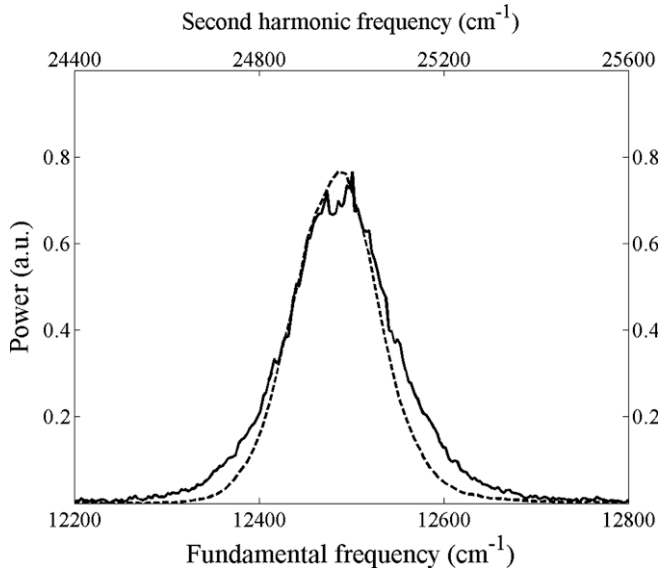

Fig. 3. Spectra of fundamental (solid line) and second harmonic (dashed line). Ocean Optics HR4000, resolution 0.75 nm.

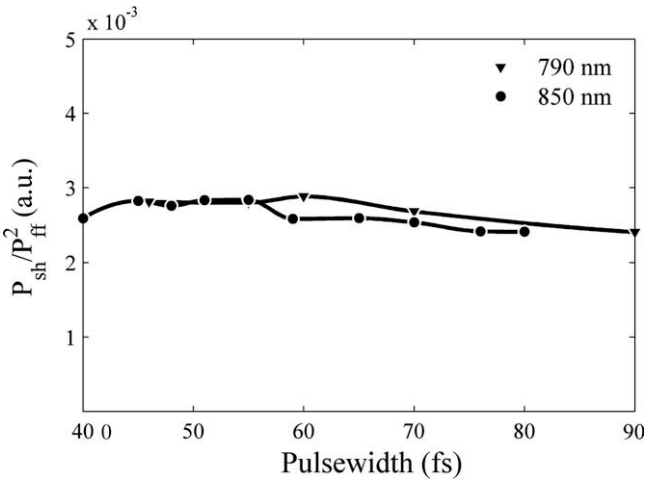

Fig. 4. Variation of second harmonic coupling coefficient with the duration of the fundamental pulses at the entrance of nonlinear medium.

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