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Phase matched conversion of radiation in nonlinear photonic crystals of strontium tetraborate

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ABSTRACT

Random quasi-phase-matched frequency doubling of fs pulses to the deep UV was obtained in 1D nonlinear photonic crystal (NPC) of strontium tetraborate. Tuning range of generated radiation is from 187.5 to 215 nm. The spectrum of generated radiation consists from series of peaks with the width of order of 1 Å. These peaks are the manifestation of the NPC band structure. Using fs oscillator as the fundamental radiation source, maximum average power of generated radiation is of order 1 μW, the enhancement factor with respect to monodomain sample being of order of several hundred. The red rotational shift of NPC band structure is experimentally demonstrated.

Keywords: nonlinear photonic crystal, strontium tetraborate, second harmonic generation, random quasi phase matching.

1. INTRODUCTION

Nonlinear optics is widely used for conversion of radiation into different spectral ranges. Conversion to deep ultraviolet (DUV) and vacuum ultraviolet (VUV) ranges is featured by limited number of nonlinear crystals transparent in these ranges as well as problems with the angular phase matching necessary for high conversion efficiency. In particular, direct phase matched second harmonic generation (SHG) in β-BaB_2O_4 (BBO) is possible down to 204.8 nm, whereas its transparency range is limited by 189 nm. Shorter wavelengths can be generated using sum frequency generation, that leads to complications of experimental setup[1]. In this case, as a rule, high power femtosecond regenerative amplifiers and parametric oscillators are used with an increased number of nonlinear crystals unavoidable necessity of delay lines for pulse synchronization. Detailed analysis of radiation generation in the vicinity of 200 nm can be found in[1].

Strontium tetraborate (SBO) nonlinear crystal is attractive from the point of view of VUV nonlinear optical generation since its fundamental absorption edge lies at the shortest wavelength among all nonlinear crystals, namely, in the vicinity of 125 nm[2]. However, in view of small birefringence, angular phase matching in SBO is absent. Due to the lack of ferroelectricity, production of regular domain structures and quasi phase matching in SBO seemed to be impossible, too. By contrast to these expectations, we have detected irregular domain structures consisting of oppositely poled domains in SBO[3]. The origin of these structures is presently unclear, and the technique to control their properties is still not developed. The refractive indices inside the structures can be considered to be homogeneous while the second order susceptibility is modulated. Therefore these structures must be classified to be a nonlinear photonic crystal (NPC)[4]. Random NPC are capable of phase matching the nonlinear conversion of broadband radiation, generally speaking, in any direction inside the crystal. Two limiting cases exist, namely, random quasi-phase-matching (RQPM) and nonlinear diffraction. From the point of view of conversion efficiency, RQPM[5] takes the intermediate position between non-phase-matched case in a monodomain sample and quasi-phase-matched case in periodically poled nonlinear crystal (PPNC)[6]. The generated power in a single domain sample oscillates along the propagation direction with the period equal to double coherence length (in absence of synchronism), while in QPM case it grows according to square law with the domain number. In random NPC the generated power experiences irregular behavior but for the interaction lengths much larger than randomization length the average linear growth of generated power will be observed[5]. Decrease in efficiency of conversion in this case is the price for the tunability in a wide spectral range. Another feature of RQPM is the preservation of conversion efficiency if the fundamental wavelength and propagation direction are increased in accordance with each other[7]. This effect can be interpreted as so-called rotational red shift of the NPC band structure.

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In the present communication we report the results of experimental study of the cascaded fourth harmonic generation of femtosecond pulses from Ti:Sapphire oscillator using RQPM in NPC of SBO at the second stage.

2. EXPERIMENTAL DETAILS

The NPC domain structure with the thickness of 2 mm along *a* crystallographic axis contained 262 domains and was well ordered in *bc* plane but highly randomized along *a* axis direction. The NPC was place on a monodomain substrate 3 mm thick in a direction. The scheme of experimental installation is presented in Fig. 1. The central wavelength of fundamental radiation was tuned in a spectral range 740-930 nm. Maximum average power of fundamental radiation was up to 960 mW. Duration of pulses was in the range 40-100 fs. Fundamental radiation was focused into the 1 mm thick doubling nonlinear crystal BBO with a ten centimeter focal length lens. Generated second harmonic radiation was collimated by means of the second ten centimeter lens. Average power of the second harmonic was up to 135 mW that corresponds to conversion efficiency of 14.4 %. Radiation of the second harmonic was selected by means of Glan prism and focused by means of a five centimeter lens into the NPC sample. The second harmonic waist radius was 37 μm on the power level $1/e^2$ that corresponds to peak power density up to 0.3 GW/cm^2. Polarization of the second harmonic coincided with crystallographic axis *c* so the maximum nonlinear coefficient d_{ccc} has been employed. The second harmonic after the NPC was suppressed with Acton 172-N bandpass filter with a maximum transmission at 173.5 nm, transmission at 200 nm being 6.5 %. Generated radiation at the frequency of the fourth harmonic was focused onto the entrance slit of MSDD 1000 monochromator equipped with matrix photodetector Hamamatsu HLS192 for registration of spectrum. The monochromator's spectral resolution in a spectral range under study was 0.023 nm (5 cm^{-1}). On an axial exit of monochromator the photomultiplier Hamamatsu H5783-04 or Newport 918D sensor operating with Newport 1931-C power meter were mounted. Maximum average power radiation of the fourth harmonic on the central wavelength of 200 nm after the NPC was 1 μW, that corresponds to conversion efficiency 10^{-5}. The generated power can be increased by two orders at least if more powerful fundamental wave source could be employed, since optical breakdown threshold of NPC material at 400 nm for fs pulses must be expected far above intensity level used in our experiment.

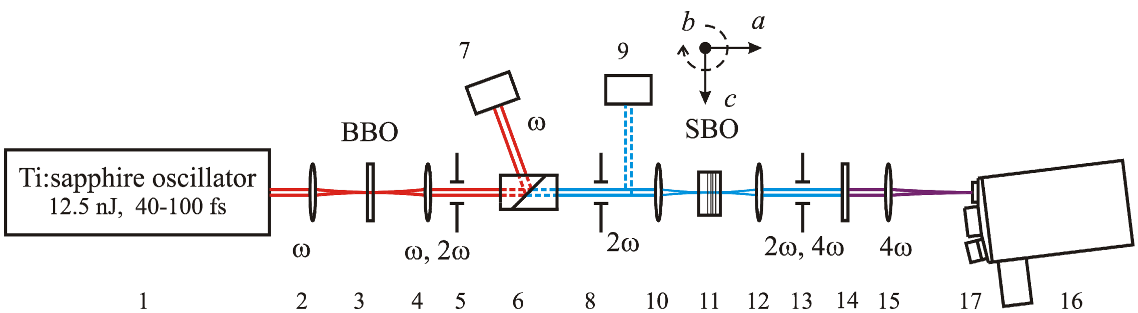

Figure 1. The experimental setup: 1 – Ti:Sapphire oscillator Tsunami/MilleniaV; 2,4 – lenses (F=10 cm), 3 – BBO crystal; 5,8,13 – an iris aperture; 6 – Glan prism; 7 – an absorber of pumping radiation; 9 – a power meter for monitoring SHG; 10,12 – lenses (F=5cm); 11 – NPC; 14 – interference filter Acton 172-N; 15 – lens (F=112mm); 16 – monochromator (Solar MSDD1000); 17 – Newport 918D sensor.

The spectra of the second and fourth harmonics after the NPC for the case of normal incidence of the input radiation onto NPC facet are presented In Fig. 2. Apparently, the spectrum of radiation of the fourth harmonic has the form not typical for a spectrum of radiation, generated in the conditions of angular phase matching. In contrast to a smooth spectrum of the second harmonic, the spectrum of the fourth harmonic radiation consists of a series of sharp peaks. Typical width of these peaks is of order of 1 Å while the distance between them lies in a range 1-3 Å. Note that the calculated spectral width of quasi phase matching in a regular domain structure optimized for frequency doubling into 200 nm would be only ~0.5 Å. Taking into consideration peak structure of the spectrum, one may expect infringement of generated radiation pulses temporal structure and increase of their duration.

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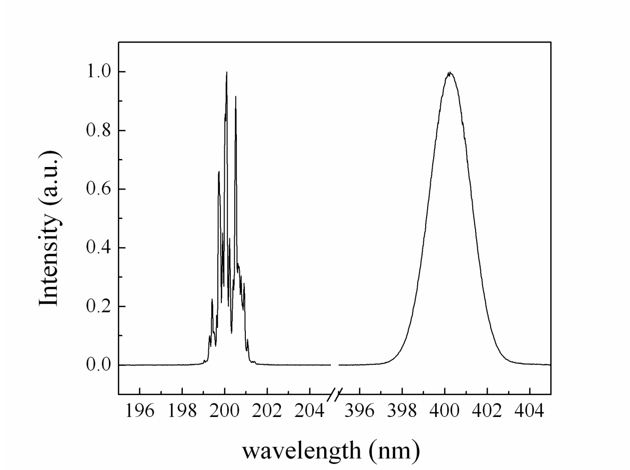

Figure 2. The spectra of the second (right) and the fourth (left) harmonics of femtosecond laser.

2.1 NPC band structure examination

In order to examine the possible rotational red shift of the NPC band structure the fourth harmonic spectra were measured as the function of incidence angle onto the NPC. The incidence angle was varied via the rotation of NPC around *b* crystallographic axis in the range from -30 to 30 degrees with the step of 1 degree. The measurement results for the generated radiation spectral intensity are presented in a contour plot (Fig.3). Really, the peaks in the fourth harmonic spectrum preserve their shape but experience shift to the longer wavelengths when NPC is rotated. This behavior allows us to identify the peak structure of the fourth harmonic spectrum as the manifestation of the NPC band structure[7]. The nature of the observed effect consists in indemnification of the interacting waves phase mismatch with increase in wavelength of the input radiation by the expense of increase in an optical thickness of domains inside the rotated NPC. Additionally, we investigated the behavior of the fourth harmonic spectrum with the variation of input radiation power and found that spectrum shape is completely independent upon this variation. This fact also testifies in favour of the NPC band structure concept. The result of NPC band structure calculation that uses the model[7] with the account for the width of fundamental radiation spectrum is presented in Fig. 3 (b). The given calculation allows to be convinced that dependence of position of spectral components on the incidence angle corresponds to the experiment.

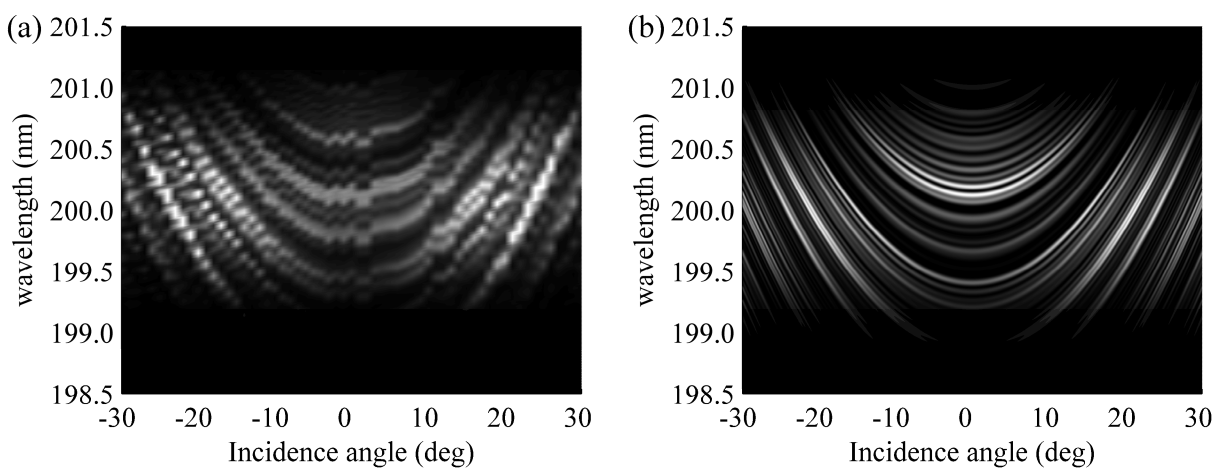

Figure 3. Experimental (a) and calculated (b) angular dependence of spectral intensity of generated radiation near 200 nm.

2.2 Enhancement factor measurement

In order to evaluate the degree of phase matching due to RQPM in our NPC sample for the conversion process under study, we compared the fourth harmonic power generated in NPC to the power obtained in a monodomain sample due to

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non-phase-matched process. The reference monodomain sample had the thickness of 432 μm (from 465 to 475 coherence lengths for various spectral components in a spectrum of the input second harmonic), with the same crystallographic orientation as the NPC. The experimental fourth harmonic spectrum from reference sample represents frequency dependence in form of “the comb” consisting from equidistant peaks under the envelope governed by spectrum envelope of input radiation (Fig. 4). The given dependence is spectral analogue of Maker’s angular oscillations[8]. Dashed line in the same figure is the calculated spectrum shape from the reference sample. The ratio of spectrally integrated powers from NPC and reference samples indicates the enhancement obtained due to RQPM. This ratio was measured to be 320 times and is close to a calculated one. However, this value is 800 times smaller than the value expected for the first order quasi phase matching enhancement factor of hypothetical regular NPC of the same thickness. Noticeable decrease in efficiency of conversion is the cost for possibility of phase matched generation of radiation in a wide spectral range.

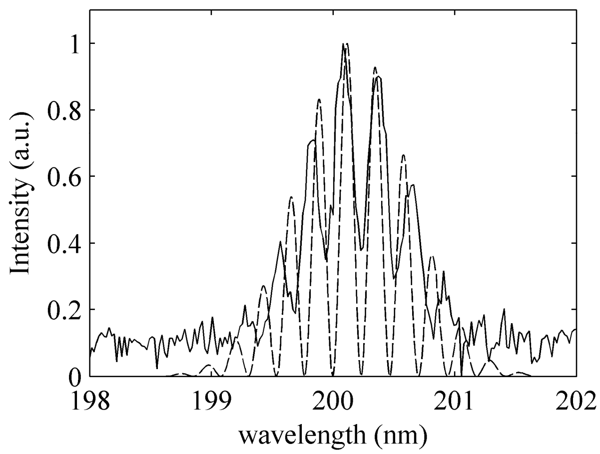

Figure 4. The measured (solid line) and calculated (dashed line) spectrum of radiation generated in the reference sample.

2.3 Fourth harmonic tunability

Tunability of the fourth harmonic (Fig. 5a) was investigated by varying the central wavelength change of the central wavelength the oscillator with the corresponding fine angular tuning of BBO doubler but with no angular tuning of NPC that was fixed in the normal incidence position. The range of tunability obtained was from 215 to 187.5 nm (Fig. 5b). The basic factor limiting the shorter wavelength cutoff is the air absorption in combination with the less important drop in the tuning curve of Ti:Sapphire laser, while absorption in NPC material produces minor impact. We have found out the peak in the absorption spectrum of our NPC sample at 209 nm with the absorption coefficient of 0.6 cm^{-1}. According to[9], absorption in this area is not caused by the basic components of NPC material. It is necessary to assume, that the given features of absorption are caused by inadvertent impurity. Hence, upon the improvement of quality of initial materials and the use of the technology providing protection of the melt from pollution in the course of growth, the given impurity can be removed.

One of features of nonlinear optical conversion of radiation in NPC is associated with a direction of radiation propagation. It is obvious, that sequences of domains on a thickness in these cases are different. However, from the point of view of nonlinear conversion these structures are equivalent since they have the same reciprocal lattice vectors spectrum. We compared the spectra of generated radiation corresponding to propagation of fundamental radiation in a forward and backward direction inside the NPC. The position of beam waist in both cases was adjusted to optimize the fourth harmonic output. As can be seen in Fig.6, the spectrum of generated radiation is independent on the direction of propagation. Beside this, it is stable upon moving the NPC on all three coordinates, including propagation direction. The stability on the transversal displacement evidences high degree of NPC ordering in the crystal plane *bc*. However, the decrease power of generated radiation by 15 % was observed when input radiation entered initially through NPC, and then through substrate. That allows to estimate the coefficient of absorption inside the substrate at 200 nm by the value of 0.54 cm^{-1}. This value is in the satisfactory consent with the absorption spectrum of NPC sample.

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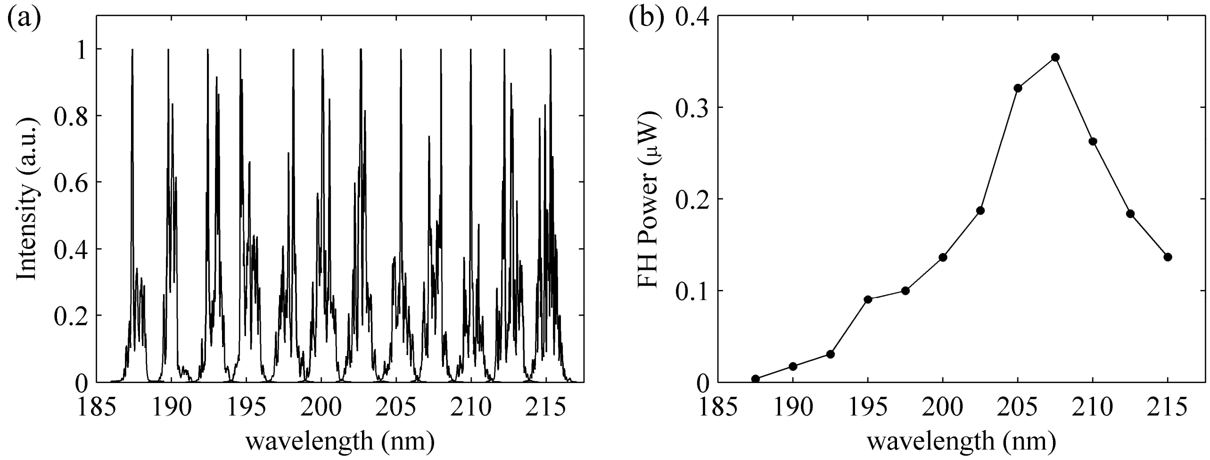

Figure 5. a) Spectra of the femtosecond laser fourth harmonic for different central wavelengths of fundamental radiation; b) Tuning curve for the average power of the fourth harmonic.

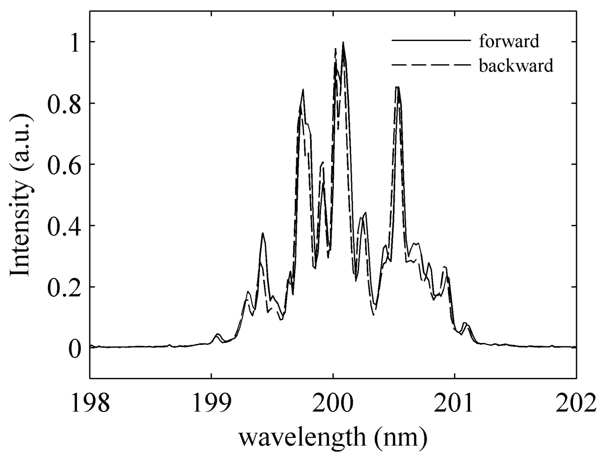

Figure 6. Spectra of generated radiation for two opposite propagation directions through NPC.

3. CONCLUSION

In conclusion, tunable generation of fourth harmonic radiation of femtosecond laser of Ti:Sapphire via RQPM in NPC of SBO in spectral range from 215 to 187.5 nm was obtained. Efficiency enhancement factor due to RQPM equals to 320 with respect to monodomain sample. Maximum average power of fourth harmonic is up to 1 μW. Spectra spiking was interpreted as the manifestation of NPC band structure. The red rotational shift of NPC band structure was experimentally observed. NPC of strontium tetraborate is capable or RQPM in deep UV and probably VUV and can be the nonlinear medium for extremely angle-nonsensitive frequency tunable converter of radiation into these spectral ranges.

4. ACKNOWLEDGEMENT

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