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Diagnostics of fs pulses by noncollinear random quasi-phase-matched frequency doubling

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Noncollinear random quasi-phase-matched frequency doubling in nonlinear photonic crystal (NPC) of strontium tetraborate (SBO) can be used for background-free autocorrelation measurements of ultrashort pulses in wide spectral range without any angular tuning of nonlinear photonic crystal. Nonuniformity of reciprocal superlattice vectors spectrum of random structure does not affect the accuracy of measurement. Nonlinear photonic crystals of strontium tetraborate are attractive medium for cross-correlation measurements in deep and vacuum ultraviolet. © 2011 American Institute of Physics. [doi:10.1063/1.3664094]

Angular phase-matched second harmonic generation (SHG) is widely used in numerous devices for autocorrelation measurements.¹ High efficiency obtainable for angularly phase-matched SHG and spatial separation of autocorrelation signal from background one allows to obtain background-free autocorrelation function for rather low power pulses. However, some problems originate from angular phase-matching concept. For instance, careful adjustment of nonlinear crystal for every value of central wavelength as well as of temporal pulse overlapping is required, which significantly complicates tuning procedure. Additionally, the edge of transparency window of the most nonlinear crystals used in pulse diagnostics devices is limited to near ultraviolet. For autocorrelation measurements at shorter wavelengths, down to 125 nm, strontium tetraborate (SBO) was used,² however, these measurements were done in the absence of any kind of phase matching, since this crystal completely lacks angular phase matching. Later, other kinds of phase matching, random quasi-phase matching (RQPM), and nonlinear diffraction, were found in SBO.^{3,4} These processes employ 1D random nonlinear photonic crystal (NPC) structure of SBO. Recently, 2D random nonlinear media based on strontium barium niobate (SBN) were studied for the purposes of pulse diagnostics.⁵⁻⁷ These experiments employ transverse emission of autocorrelation signal that is in fact the limiting case of nonlinear diffraction geometry. The signal-to-noise ratio in this geometry was found to be rather low, of order of 4.⁵ Random NPCs in SBO were investigated for pulse diagnostics in Ref. 8, and in this case, so-called nonlinear diffraction from virtual beam⁹ was used. The contrast of order of several thousands, good accuracy of pulse duration measurement and insensitivity of the nonlinear crystal to the central wavelength of the pulse were reported in Ref. 9. RQPM, also potentially applicable for pulse diagnostics, was not investigated up to date, since pure case of RQPM can be achieved only in 1D random media. Recently, large potential of random 1D NPC in LiNbO₃ was demonstrated in Ref. 10, where conversion efficiencies above 20% were obtained for the

generated wavelengths in the red to green spectral regions. In the latter paper, picosecond pulses of fundamental radiation were employed. However, the peaky structure of generated radiation spectrum found for the case of RQPM in NPC SBO,¹¹ where femtosecond fundamental pulses were used, may affect the accuracy of fs pulse diagnostics, since it is generally recognized that for accurate measurements, all the spectral components must be uniformly converted.¹²

In this letter, we report the study of noncollinear random quasi-phase matched frequency doubling in nonlinear photonic crystal of strontium tetraborate and its employment for background-free autocorrelation measurements of ultrashort pulses in wide spectral range without any angular tuning of nonlinear photonic crystal. In our experiment, we used common autocorrelation scheme with adjustable delay line to control overlapping between two fundamental radiation pulses from a femtosecond oscillator (Tsunami, Spectra-Physics) operating at the repetition rate 82 MHz. The sample under study was the crystal of strontium tetraborate with the dimensions $5 \times 11 \times 9$ mm³ along the crystallographic axes a , b , and c (in a non-standard $Pnm2_1$ space symmetry group). All facets of the sample are perpendicular to corresponding crystallographic axes. The sample contained a sequence of naturally grown domains with the domain walls perpendicular to the crystallographic axis a , which form 1D NPC. Length of NPC in a axis direction was 2 mm, while residual 3 mm of sample were a monodomain substrate. In the autocorrelation measurements, the laser beams entered through input facet of NPC but not through that of the substrate, in order to avoid any unnecessary temporal spread of the pulses due to group velocity dispersion (GVD).

Fundamental beams are focused into NPC by 10-cm cylindrical lens [Fig. 1(a)] with the external crossing angle approximately 7°. The bisector of intersecting angle of two fundamental beams was perpendicular to the input facet of SBO. Polarization of fundamental radiation was vertical and coincided with crystallographic axis c in order to employ the largest nonlinear coefficient d_{ccc} of SBO. Average power of each fundamental beam was equal to 340 mW. Each of fundamental beams generated collinear second harmonic radiation independent on delay. Average second harmonic

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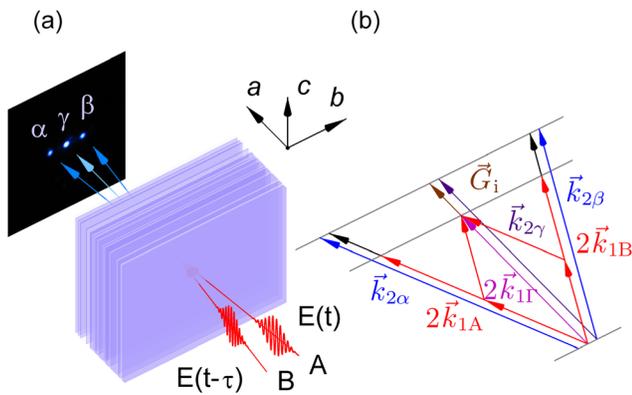


FIG. 1. (Color online) Experimental layout of autocorrelation measurements (a) and corresponding phase-matching diagram (b).

power was $0.4 \mu\text{W}$ in each of these side beams. When both fundamental pulses are temporally overlapped inside the NPC, additional noncollinear second harmonic beam (denoted by γ) appears, as presented in Fig. 1(a). Origin of central beam can be understood from vectorial phase-matching diagram presented at Fig. 1(b). Momentum conservation for second harmonic generation in the side beams is $\vec{k}_{2\alpha} = 2\vec{k}_{1A} + \vec{G}_i$ and $\vec{k}_{2\beta} = 2\vec{k}_{1B} + \vec{G}_i$, while for the central beam $\vec{k}_{2\gamma} = 2\vec{k}_{1\Gamma} + \vec{G}_i$, where \vec{k}_{1A} , \vec{k}_{1B} , $\vec{k}_{1\Gamma}$ and $\vec{k}_{2\alpha}$, $\vec{k}_{2\beta}$, $\vec{k}_{2\gamma}$ are the fundamental and second harmonic wavevectors, respectively, and \vec{G}_i represents a vector from an infinite set of NPC reciprocal superlattice vectors. Effective fundamental wavevector, which is responsible for the noncollinear conversion process, is $\vec{k}_{1\Gamma} = 1/2(\vec{k}_{1A} + \vec{k}_{1B})\cos(\theta)$, where 2θ is intersecting angle of fundamental beams. Intensity of central beam is determined by delay and position of focal plane inside the NPC. To maximize the output of second harmonic power, we adjusted the maximum overlapping of two noncollinear fundamental beams and then adjusted the position of NPC along a axis, maximized average power of autocorrelation signal being $1.9 \mu\text{W}$. In spite of both side beams and central beam arise from random quasi-phase matching, the spectra of them significantly differ. As can be seen from Fig. 2, SH spectrum of central beam is much more smooth than that of SH spectrum in a side beam. The full width at half maximum (FWHM) of autocorrelation signal is 3.08 nm in NPC SBO. The smoothness of autocorrelation signal spec-

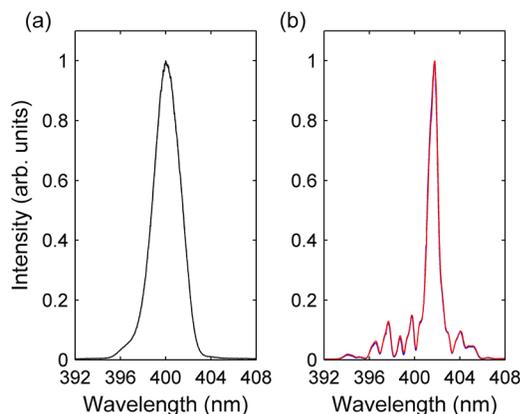


FIG. 2. (Color online) Second harmonic spectra of autocorrelation signal (a) and of single beams (b) obtained using NPC SBO. Both single-beam SH spectra coincide each other.

trum can be explained by the action of three factors: overlapping the two pulses with fs duration in time, overlapping of the crossing beams in space, and by the different range of \vec{G}_i vectors contributing to the autocorrelation signal. The latter factor, however, must not radically influence the spectra shape. The influence of first two factors on the SH intensity was discussed in Ref. 8, but their influence on the SH spectra was not investigated.

In autocorrelation measurements, we used 0.5 mm thick beta barium borate crystal (BBO) as a reference. This reference crystal was cut at angular phase matching angle for 800 nm . The average second harmonic power was measured by using Newport (Irvine, CA) 918UV/1931 C detector with BG39 filter installed in front of it. We used additional OD1 filter before BG39 in order to attenuate second harmonic intensity generated in BBO to prevent two-photon absorption in BG39 filter. Average second harmonic power of the central beam generated in BBO was $896.5 \mu\text{W}$, the FWHM of spectrum being 3.63 nm . Varying the delay between input fundamental beams results in autocorrelation function presented in Fig. 3 in a linear scale (b) and in a logarithmic one (c). Good agreement between autocorrelation traces is evident. While determining the pulse duration from autocorrelation traces, we used Gaussian temporal shape of the pulses that gives better fitting than hypersecant one. The pulse

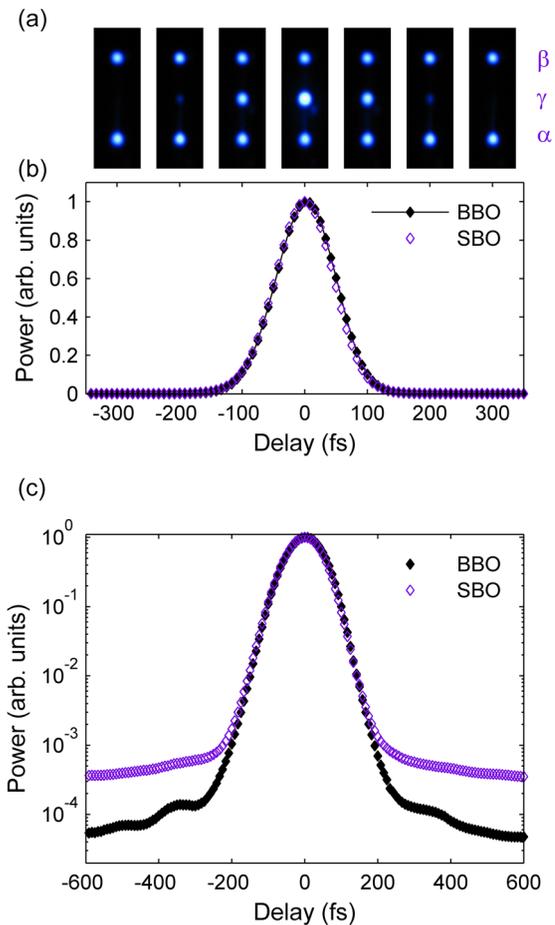


FIG. 3. (Color online) (a) Pattern of the beams with changing delay. (b) and (c) Autocorrelation functions measured using NPC SBO ($\tau = 77.9 \text{ fs}$) and 0.5 mm thick BBO ($\tau = 79.6 \text{ fs}$) in a linear (b) and logarithmic (c) scales. Attached movie demonstrates nonlinear interaction pattern upon delay (enhanced online) [URL: <http://dx.doi.org/10.1063/1.3664094.1>].

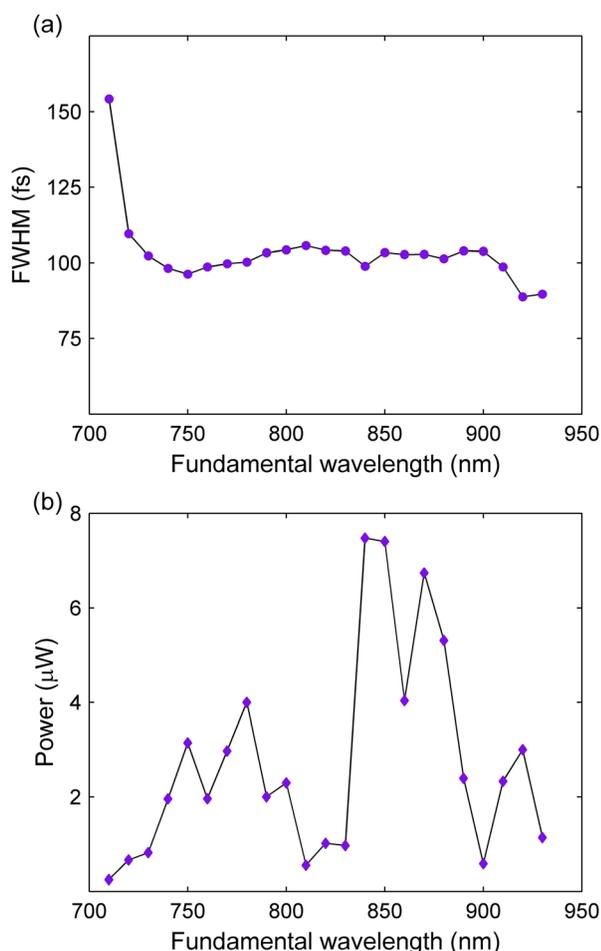


FIG. 4. (Color online) (a) Pulse duration measurement in the tuning range of Ti:Sapphire oscillator at fixed angular position of NPC. (b) Power of autocorrelation signal in the course of pulse duration measurement.

duration measured using NPC SBO was 77.9 fs, while that measured with BBO was 79.6 fs. Calculated GVD for SBO is approximately $40.8 \text{ fs}^2 \cdot \text{mm}^{-1}$ that more than 2 times lower than GVD for *ooe*-type of interaction in BBO. However, in both nonlinear media expected broadening of pulses under measurement is of order of 0.8 and 0.5 fs, correspondingly, and can be neglected. Signal-to-noise ratio is 2800 for SBO and 19300 for BBO. This ratio is not only much better than for autocorrelation experiments with other random media,⁵ but also exceeds that obtained with NPC SBO in geometry of nonlinear diffraction from virtual beam.⁸ However, we think that in the latter case, the difference is not principal and can be changed via tuning the optical scheme.

Similar autocorrelation measurements were performed for all accessible tuning range of femtosecond oscillator from 710 to 930 nm with 10 nm central wavelength steps. Fig. 4(a) presents the results of this measurement performed without any rotation of NPC. Despite rather strong variation

of the autocorrelation signal power during this measurement that arises from nonuniformity of \vec{G}_i vectors spectrum of NPC sample under study (Fig. 4(b)), stable and accurate measurement is performed throughout all investigated tuning range. Variation of pulse duration observable in Fig. 4(a) surely reflects variations in fs oscillator tuning but not the variations in measurement procedure.

In conclusion, we have shown, that random quasi-phase matched frequency doubling in nonlinear photonic crystal of strontium tetraborate may be employed for background-free autocorrelation measurements of ultrashort pulses in wide spectral range without any angular tuning of nonlinear photonic crystal. Nonuniformity of reciprocal superlattice vector spectrum both at the scale within the pulse bandwidth and at the scale of the whole tuning range of a laser does not affect the accuracy of measurement. The results of measurement are insensitive to the displacement of NPC along the axis or high randomization. Nonlinear photonic crystals of strontium tetraborate are also attractive medium for cross-correlation measurements in deep and vacuum ultraviolet and for simplest calibration of optical schemes in pump-probe experiments.

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