# Random spectrally resolved Maker fringes

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Received May 24, 2013; revised June 27, 2013; accepted June 28, 2013; posted June 28, 2013 (Doc. ID 191168); published July 23, 2013

The Maker fringes technique is extended to the case of nonlinear media with one-dimensional second-order nonlinear susceptibility modulation. For broadband radiation, second harmonic intensity oscillates in both spectral and angular domains, which can be considered random spectrally resolved Maker fringes. A theoretical approach is developed for modeling the second-harmonic generation in such domain structures, and the calculations are in excellent agreement with experimental results. 

*OCIS codes:* (190.2620) Harmonic generation and mixing; (190.4223) Nonlinear wave mixing; (160.4330) Nonlinear optical materials.

http://dx.doi.org/10.1364/OL.38.002691

Second-harmonic generation (SHG) is the most studied nonlinear-optical phenomenon that allows one to extend the spectral range of common laser sources to shorter wavelengths. There are two widespread techniques to achieve efficient SHG by matching the phase velocities of the two waves, which are based on crystal birefringence or on structuring nonlinear media known as nonlinear photonic crystals (NPCs) [1]. The latter relies on reciprocal lattice vectors (RLVs) to compensate for the phase mismatch between the interacting waves. In simplest periodic structures, quasi-phase-matching is realized [2]. At present, periodic NPCs are widely used in both science and technology, while aperiodic and random NPCs are of special interest because of their broad RLVs spectrum [3]. In general, such structures employ random quasi-phase matching (RQPM) [4–7] and can be used for cascaded harmonic generation [8], pulse compression and shaping [9], ultrashort pulse diagnostics [10,11], etc. So far, many types of random 1D NPCs have been fabricated, such as aperiodic [12,13], chirped [14], and Fibonacci-like [15], but at the same time, no as-grown random 1D NPCs are known except for strontium tetraborate (SBO). The quadratic nonlinear susceptibility of such NPCs based on SBO stochastically alternates as a function of a single coordinate, leading to an ultrabroad spectrum of the RLVs that enables compensation of phase mismatch of various frequency-mixing processes over extremely wide spectral and angular ranges [16]. For quasi-monochromatic radiation, the angular dependence of second-harmonic intensity due to RQPM [17] is found to be in close analogy to the well-known Maker fringes observed in a single crystal plate away from phase matching. When broadband femtosecond pulses are used as fundamental radiation, spectrally resolved Maker fringes (SMFs) are observed in thin crystal plates [18,19]. These SMFs represent a frequency comb with a smooth envelope. In the case of random NPCs and femtosecond pulses, the SMFs must be modified. In this Letter we report our study of SMFs in the process of SHG of femtosecond pulses in a random 1D SBO NPC.

In the experiments, an SBO sample with dimensions $5\ \mathrm{mm} \times 11\ \mathrm{mm} \times 9\ \mathrm{mm}$ along $a$, $b$, and $c$ axes, respectively, was used. The opposite polar faces ($c^+$ and $c^-$ planes) of the sample were polished and etched to visualize the NPC structure [20]. The domains were measured by using optical Axio Observer.A1m (Carl Zeiss, Inc) and electron TM-1000 (Hitachi) microscopes. Figure 1(b) shows an optical image of a part of the etched pattern of the NPC structure. The domain structures from two opposite faces were analyzed and compared with each other. The domain sequences from opposite faces are found to be in excellent agreement, which verifies the good uniformity of NPC structure in the $bc$ plane. The domain structure contains 275 domains with overall thickness ~2360 μm, so that the mean value of domain thickness is 8.6 μm. The distribution of domain thicknesses is asymmetric with respect to the maximum at 0.4 μm, with a wide wing on the side of thick domains, resulting in the standard deviation 20.1 μm. The part of the sample 2.64 mm thick in the $a$ axis direction was a single domain and can be treated as a substrate. The sample under study was mounted on a rotation stage. Fundamental radiation from a femtosecond Ti:sapphire oscillator (Tsunami, Spectra-Physics) was focused into the NPC by a 10 cm focal lens ($L_1$) along the SBO $a$ axis [Fig. 1(a)]. It was polarized in the vertical plane parallel to the crystallographic $b$ axis so that the nonlinear coefficient $d_{cbb}$ was employed.

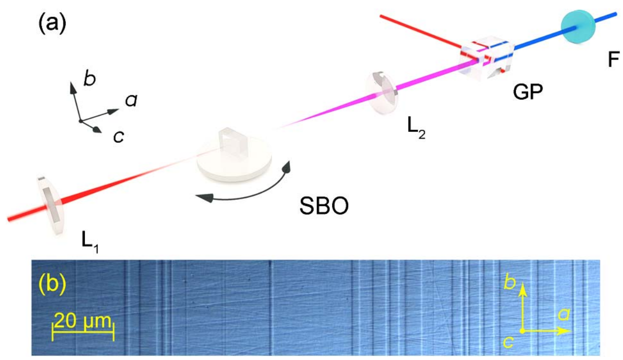


Fig. 1. (a) Experimental scheme. (b) Part of etched face of SBO normal to the $c$ axis.

 

The fundamental (FF) and the generated second harmonic (SH) were collimated by a second 10 cm lens ($L_2$), and FF was separated by Glan prism (GP) and absorbing filter (F). The SH spectra were measured by using a spectrometer (MSDD1000, Solar TII) with a spectral resolution of 0.02 nm.

A simple theoretical model for the SHG of femtosecond pulses in a random NPC can be developed by using the approach from [21]. Under the slowly varying amplitude approximation and in the low conversion limit, the amplitude of the SH generated within a single domain of thickness $z$ can be represented in the most general form as $A(\Omega, z) \propto \alpha E \otimes E'(\exp[i(\Delta k - \nu\Omega)z] - 1)$, where $\alpha = -id_{cbb}\omega_1^2/(k_2 c^2(\Delta k - \nu\Omega))$, $E \otimes E'$ is the convolution of the fundamental spectrum, $\Delta k = 2k_1 - k_2$ is the wave vector mismatch for FF ($\omega_1$) and SH ($\omega_2$) waves, $\nu$ is the group-velocity mismatch and $\Omega = \omega_2 - 2\omega_{10}$ is the frequency detuning, with $\omega_{10}$ being the central frequency of the FF wave. The product $\nu\Omega$ governs the walk-off between FF and SH pulses owing to the group-velocity mismatch. For a Gaussian spectral shape of the FF field with full spectral width $2\Delta\Omega$, the convolution factor is $E \otimes E' = \sqrt{\pi/2}\exp(-\Omega^2/2\Delta\Omega^2)$. Consequently, the spectral amplitude of the SH field after the first $N$ domains is expressed as follows:

$$A(\Omega, \theta) = \sqrt{\frac{\pi}{2}}\alpha\xi(\theta)\exp\left(-\frac{\Omega^2}{2\Delta\Omega^2}\right) \times \left(\sum_{n=1}^{N}(-1)^n\left(\exp\left[i\frac{(\Delta k - \nu\Omega)d_n}{\cos\theta}\right] - 1\right) \times \exp\left(i\Delta k\sum_{r=n+1}^{N}\frac{d_r}{\cos\theta}\right)\right) \quad (1)$$

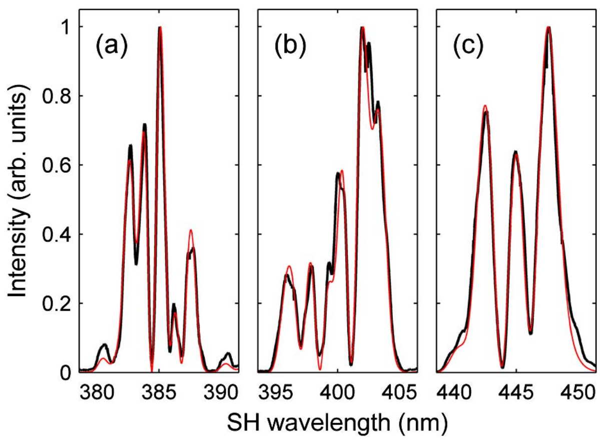


Fig. 2. Measured (black) and calculated (red) SH spectra for different central fundamental wavelengths: (a) 770 nm, (b) 800 nm, and (c) 890 nm.

with $d_n$ being the thickness of $n$th domain and $\xi(\theta)$ denoting an envelope factor for the $mm2$ point group crystals, which can be found in [22]. The factor $\exp(i\Delta k\sum_{r=n+1}^{N} d_r/\cos\theta)$ accounts for the phase accumulated from the exit plane of the $n$th domain to the exit plane of the $N$th domain. The factor $\cos^{-1}\theta$ takes into account the dependence of the optical path on the angle of FF propagation inside the sample. The SH spectral intensity is given by $S(\Omega, \theta) = |A(\Omega, \theta)|^2$. Refractive index data were taken from [23].

As one can see in Fig. 2, the measured SH spectra are structured as expected for RQPM-type SHG under pumping by broadband radiation. The calculated SH spectra $S(\Omega, \theta = 0)$ are in good agreement with the experimental ones. For the calculation, the input parameters were fitted by using a least-mean-square algorithm. The necessity of fitting the wave vector mismatch $\Delta k$ might be caused by instrumental inaccuracy of the microscope used and by refractive index uncertainties. The experimental and computed values of parameters that underwent fitting are presented in Table 1. The product $\nu\Omega$ was omitted in the calculations, having in mind that under the experimental conditions ($\nu = 216$ fs/mm) the mean value of domain thickness $\langle d\rangle$ is smaller than the group walk-off length ($\langle d\rangle \ll L \cong 370$ μm). We conclude that the same results can be obtained with, e.g., nanosecond pulses with wavelength tuning, though durable measurement would be necessary in that case. So, the role of the femtosecond laser in the present experiment is only the delivery of broadband radiation that makes measurements easier.

The measured SH spectrum exhibits pronounced angular dependence in the range of NPC rotation angle from 0 to 30 deg as depicted in Fig. 3(a). In analogy with well-known angular Maker fringes [17] and their spectral extension observed in nonlinear crystal plate away from phase matching [18], we can generalize these features for the case of random 1D media when the measured dependence can be considered as random SMFs. The main feature of these SMFs is that the spectral peaks shift toward longer wavelengths within the spectral envelope ascribed to the fundamental radiation with NPC rotation angle. According to Eq. (1), the origin of this shift can be understood in sense of $\Delta k(\Omega)/\cos\theta = \text{constant}$. Figure 3(b) shows that the calculated spectral SH intensity $S(\Omega, \theta)$ behaves in the same manner as the measured one, and they are in excellent agreement. While analysis of common Maker fringes provides important information about the coherence length and the nonlinear coefficients involved, extraction of such information in the case of broadband pulses and random media is more complicated. It requires complete knowledge of the thickness of the individual domains. Therefore, SMFs affected by a random domain structure

**Table 1. Experimental and Fitted Values of Parameters**

| | Experimental | | Fitted | | |
|---|---|---|---|---|---|
| Fundamental Wavelength (nm) | CWL (nm) | FWHM (nm) | CWL (nm) | FWHM (nm) | $\Delta k_{fit}/\Delta k$ |
| 770 | 769.9 | 10.8 | 771.0 | 11.1 | 1.0175 |
| 800 | 800.2 | 10.5 | 800.5 | 11.2 | 1.0181 |
| 890 | 890.1 | 10.5 | 890.1 | 11.4 | 1.0165 |

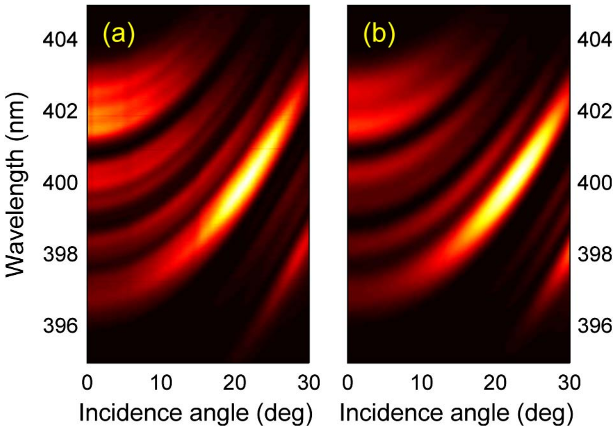


Fig. 3. (a) Measured and (b) calculated random SMFs.

are less informative compared to a single crystal. Nevertheless, integration of SMFs over the spectrum leads to smooth angular dependence of SH intensity, resulting in ultranoncritical SHG with enhanced intensity.

The good agreement between experimental and theoretical results allows one to use Eq. (1) for computation of the evolution of the SH spectral components in the domain structure. Figure 4 shows SH spectra calculated at domain wall positions for forward and backward propagation, demonstrating the decomposition and narrowing of spectral peaks with the propagation distance. In spite of the fact that the evolution of SH spectra along opposite directions is rather different [Fig. 4(b) versus 4(c)], the resulting SH spectrum is independent of the propagation direction. This is direct evidence that the set of RLVs plays the main role in realization of the frequency conversion processes, and both sequences of domains are identical with respect to their nonlinear features. A difference between forward and backward SH spectra is expected only if the group-velocity mismatch effects and absorption of the material are large enough.

In our experiments, the SH radiation was continuously tuned in the spectral range 355–470 nm. The maximum average SH power of ~50 μW was obtained at a wavelength of 390 nm. The SHG efficiency attained was $10^{-4}$ which is a typical value for highly randomized

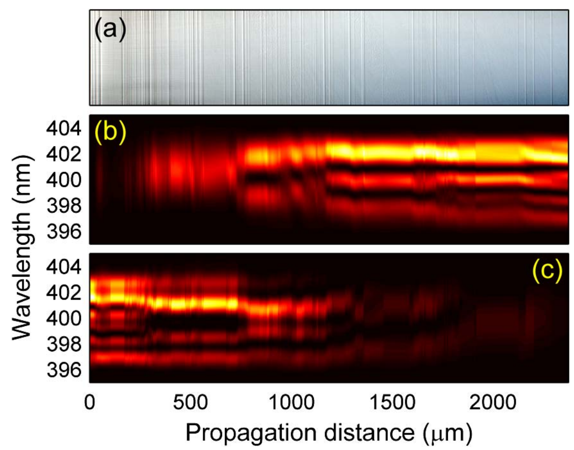


Fig. 4. (a) Domain pattern and evolution of the SH spectrum as a function of the propagation distance in (b) forward and (c) backward directions.

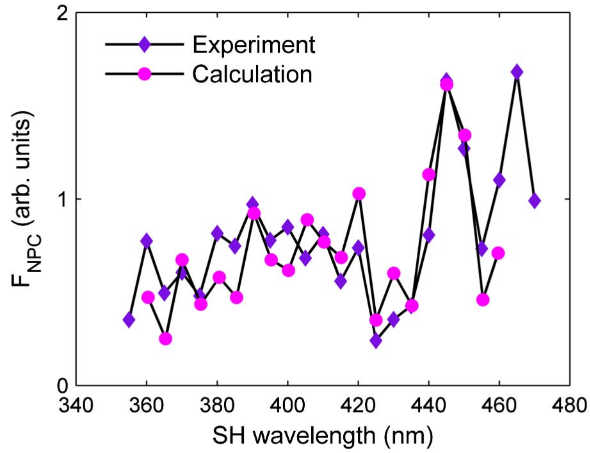


Fig. 5. Measured and calculated tuning curves of the averaged SH power divided by the squared FF power.

nonlinear media. The spectral dependence of the averaged SH power divided by the squared FF power $F_{\mathrm{NPC}} = P_{2\omega}/P_{\omega}^2$ is shown in Fig. 5. This dependence is not influenced by the FF intensity and reflects the contribution of the domain structure to the SHG. Calculated points in Fig. 5 were obtained by using parameters fitted to obtain good agreement between calculated and measured spectra of SH as demonstrated in Fig. 2 for three specific wavelengths. As a result, good agreement between experimental and calculated data is also observed in Fig. 5.

In conclusion, we studied random quasi-phase-matched SHG of femtosecond pulses in a 1D SBO NPC. The Maker fringes technique is extended to the case of 1D nonlinear media with second-order nonlinear susceptibility modulation. For broadband radiation, the SH intensity oscillates in both its spectral and angular dependence, which can be interpreted as random SMFs. Calculations are in excellent agreement with experimental results. SHG generated under RQPM is characterized by a spectrum distorted with respect to the fundamental. This feature, however, may create new possibilities for ultrashort pulse shaping in coherent control.

The work was partially supported by the grant of the President of the Russian Federation MK-250.2013.2, by RFBR through grant 12-02-31167, by the Ministry of Education and Science of Russian Federation (contract 14.B37.21.0730), by the Krasnoyarsk Regional Fund of Science and Technical Activity Support, by PSB RAS Project 2.5.2, and by SFU grant F12.

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