Structural light focusing phenomenon and enhanced second harmonic generation in NaNO$_2$-infiltrated opal photonic crystal

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Abstract. We report new experimental results on enhanced second harmonic generation using a structural light focusing phenomenon in photonic crystals (PCs). We use opal-based PC, infiltrated with NaNO$_2$ and pumped with femtosecond laser pulses at various incidence angles, in order to examine the dependence of second harmonic generation efficiency on the pumping wavelength location toward the PC band-gap. We demonstrate one order enhancement of second harmonic generation in case of PC band-gap pumping in comparison to non-band-gap pumping. Second harmonic generation is performed in reflection mode with the maximum of generation in the direction of mirror reflection. We demonstrate that the spectrum of second harmonic does not narrow with the quasi-phase matching condition in case of band-gap generation, and second harmonic spectrum corresponding to non-band-gap generation undergoes 1.5 times narrowing due to the quasi-phase matching.

1. Introduction
The problem of highly efficient high optical harmonic generation are of high importance in photonics, nonlinear optics, and laser physics. Photonic band-gap (BG) structures, photonic crystals (PC) and quasi-crystals, metamaterials have attracted considerable interest as a possible media for this purpose [1–12]. The PC is a structure characterized by a spatially periodic distribution of medium optical properties, and this feature of PCs leads to appearance of numerous physical effects, including nonlinearity of dispersion relation [1–4] and optical field localization [10–12]. PCs are novel media for nonlinear optics and laser physics.

Many papers are dedicated to enhancement of nonlinear light conversion in a PC on the basis of fulfillment of the quasi-phase matching condition utilizing nonlinearity of PC dispersion relation $q(\omega)$. In paper [6] the condition for enhance sum-frequency generation is examined by means of tuning the spectral position of the two mixing wavelengths near the PC band-gap. In manuscripts [7–9] enhanced nonlinear conversion is produced by compensation of the phase mismatches between the fundamental and the second (or third)-harmonic waves, when the harmonic wavelength was tuned across the band-gap of the colloidal PC, $\lambda_{2\omega}/3\omega \sim \lambda_{BG}$, and the fundamental wavelength was far red-shifted, $\lambda_{\omega} \gg \lambda_{BG}$. Papers [10–12] introduces another mechanism for nonlinear conversion enhancement in PSs based on structural light focusing effect, which leads to strong optical field localization near the PC surface in case of band-gap pumping. Problem of experimental observation and theoretical description of nonlinear
conversion enhancement via the structural light focusing in PCs and quasi-PCs [13] is of high importance for the development of novel nonlinear media.

This paper presents novel experimental results of studying the second harmonic generation enhancement via the structural light focusing phenomenon in PCs. Second harmonic generation is experimentally observed in NaNO$_2$-infiltrated opal PC pumped with the femtosecond optical pulses, and the dependence of second harmonic intensity on incidence and reflection angles, $\theta_I = \theta_R$, and, as a consequence, on fundamental pumping wavelength, $\lambda_\omega$, position towards the PC band-gap, $\lambda_{BG}$, is studied. The highest second harmonic intensity corresponds to $\lambda_\omega = \lambda_{BG}$, and almost one-order enhancement of the second harmonic generation is experimentally achieved in this case in comparison to non-band-gap PC pumping, $\lambda_\omega \gg \lambda_{BG}$. The physical origin of observed phenomenon is associated with structural light focusing effect, leading to strong optical field localization and highly efficient PC pumping.

2. Experimental results

We have used opal globular PC, consisting of fused silica microspheres having $d \approx 400$ nm diameter, and made of colloid solution, as a basis for nonlinear PC manufacturing. Fused silica microspheres were close-packed in a face-centered cubic (FCC) lattice, and the voids between globules were infiltrated with melted NaNO$_2$, having lower melting temperature than the fused silica. Figure 1 shows the schematic representation of the PC and the process of nonlinear light scattering. Poly-domain NaNO$_2$ crystalline structures, possessing non-zero quadratic susceptibility $\hat{\chi}^{(2)} \neq 0$ [14] were formed in the voids after the PC cooling. Sample manufacturing and PC void full-filling were controlled by means of scanning electron microscopy. Fused silica globules in (1 1 1) PC layer possess regular hexagonal arrangement with 0.9 factor of void full-filling. The (1 1 1) layers of the PC are periodic along the growing direction, thus, the light is assumed propagating along or close to the [1 1 1] direction of the structure. The result of the PC band-gap characterization is presented in the figure 2 and corresponds to the incidence and reflection angles $\theta_I = \theta_R = 10^\circ$: sharp band-gap centered at $\lambda_{BG} = 980$ nm exists.

We have utilized the femtosecond Yb:KGW laser with a linear polarized output to pump the sample at various angles of incidence $\theta_I$. Fundamental wavelength of femtosecond laser was $\lambda_\omega = 1026$ nm; pulse duration was $\tau \approx 250$ nm; pulse repetition rate was $f = 200$ kHz; and average power was 3.6 W. Second harmonic polarization was controlled with a dichroic

![Figure 1. Schematic representation of nonlinear light scattering on a PC. PC filler is not presented in a scheme for simplicity.](image-url)
Figure 2. Reflectivity of NaNO$_2$-infiltrated opal PC corresponding to the incidence and reflection angles $\theta_I = \theta_R = 10^\circ$. The peak at this curve corresponds to PC band-gap, $\lambda_{BG} = 980$ nm.

film polarizer to be orthogonal to the plane of incidence. Therefore, $s$-polarized fundamental wave radiated the sample, and $s$-polarized second harmonic wave was detected. Laser beam was slightly focused on the PC surface, thus, $D = 2$ mm beam spot diameter was achieved. After nonlinear light scattering the spectrum of the $s$-polarized optical field intensity was detected at far-field utilizing the diffraction grating spectrometer and the optical fiber probe. Since both the sample and the probe were placed on motorized rotational stages, incidence and reflection angles could be varied, and nonlinear scattering indicatrix could be measured. The fiber probe is used in order to maximize the resolution of the SHG indicatrix characterization.

Figure 3 shows the spectrum of the $s$-polarized second harmonic corresponding to the incidence and reflection angles $\theta_I = \theta_R = 10^\circ$. In this case the band-gap pumping condition, $\lambda_\omega = \lambda_{BG}$, is satisfied, and highest second harmonic intensity is observed. It is in complete agreement with the results of numerical simulations performed in [11]. In [11] the enhancement of second harmonic generation was predicted for the case of pumping wavelength matches the PC band-gaps

$$\lambda_\omega = \lambda_{BG_i}, \quad i = 1, 2, ..., N.$$

Almost 2\% conversion efficiency was reached for the band-gap pumping: $\approx 70$ mW power of $s$-polarized second harmonic, radiated in the direction of mirror reflection and having approximately $\pm 10^\circ$ divergence, was measured utilizing the power meter.

With an increase of $\theta_I = \theta_R$ the PC band-gap, $\lambda_{BG}$, undergoes blue-shift [9,12]

$$\lambda_{BG} \approx 2d\sqrt{\tilde{n}^2 - \sin^2(\theta)},$$

where $\tilde{n}$ is an effective refractive index of PC structure

$$\tilde{n} = \sqrt{(1 - f)n_{SiO_2}^2 + fn_{NaNO_2}^2},$$

$n_{SiO_2}$ is a refraction index of fused silica, $n_{NaNO_2}$ is a refraction index of NaNO$_2$ voids, and $f$ is NaNO$_2$ volume fraction. Therefore, the dependence of second harmonic generation efficiency on the fundamental wavelength location, $\lambda_\omega$, toward the band-gap, $\lambda_{BG}$, could be examined by rotating the PC. Figure 4 shows the decreasing of the second harmonic generation intensity with growth of equal pair of incidence and reflection angles, $\theta_I = \theta_R$. One order enhancement of the
second harmonic generation was experimentally observed for the band-gap pumping, \( \lambda_\omega = \lambda_{BG} \) (\( \theta_I = \theta_R = 10^\circ \)), in comparison to non-band-gap pumping, \( \lambda_\omega > \lambda_{BG} \), (\( \theta_I = \theta_R > 10^\circ \)).

Figure 5 shows the dependence of second harmonic spectrum width at full-width at half maximum, FWHM\(_{2\omega}\), towards the increasing angles, \( \theta_I = \theta_R \). Strong decrease of FWHM\(_{2\omega}\) is observed with \( \theta_I = \theta_R \) increasing, and, as a consequence, with \( \lambda_{BG} \) blueshifting. This curve confirms, that nonlinear generation occurs in the thin near-surface area of the PC in case of the band-gap pumping, \( \lambda_\omega = \lambda_{BG} \) [11,12], and it performs in the PC volume in opposite case, \( \lambda_\omega \neq \lambda_{BG} \) [7–9]. This narrowing of second harmonic spectrum is associated with the quasi-phase

Figure 4. Dependence of second harmonic intensity, \( I_{2\omega} \), on \( \theta_I = \theta_R \). Varying \( \theta_I = \theta_R \) we are examining the dependance of second harmonic generation efficiency on the ration \( \lambda_\omega/\lambda_{BG} \), and the most efficient second harmonic generation corresponds to PC band-gap pumping, \( \theta_I = \theta_R = 10^\circ \) and \( \lambda_\omega = \lambda_{BG} \).
Figure 5. Dependence of second harmonic spectrum width, $FWHM_{2\omega}$, on $\theta_I = \theta_R$. The most wideband second harmonic generation corresponds to PC band-gap pumping, $\theta_I = \theta_R = 10^\circ$ and $\lambda_\omega = \lambda_{BG}$, since no quasi-phase matching condition [7–9] should be satisfied for the band-gap generation case [12].

matching condition, which should be satisfied in case of generation occurs in PC volume [7–9].

One could perform efficient second harmonic generation in PC volume if the condition of effective nonlinear diffraction [7-9,12] is satisfied

$$k_{2\omega} = k_\omega + nG_{111}$$

where $k_\omega$ is a fundamental wave vector, $k_{2\omega}$ is a second harmonic wave vector, $G_{111}$ is a reciprocal vector of the FCC lattice in [1 1 1] direction, and $n$ is an order of the PC band-gap. This condition takes into account interference between the SH fields from all NaNO$_2$ infiltrated voids, and the interference is constructive only for thin bandwidth of the second harmonic spectrum. In case of PC band-gap pumping, $\lambda_\omega = \lambda_{BG}$, second harmonic generation is performed in several near-surface layers of PC [10–12], and condition (4) becomes insignificant, since second harmonic field at far-field region is a superposition of the second harmonic fields generated in the NaNO$_2$-infiltrated voids of several near-surface PC layers. Thus, no narrowing of second harmonic spectrum is observed for the band-gap generation [11,12] in contrast to the previous quasi-phase matching approaches [7–9]. Strong optical field localization due to the structural light focusing and no spectrum narrowing are main advantages of considered technique of second harmonic generation in a PC.

3. Discussions

Observed second harmonic generation demonstrates both an existence of second-order nonlinearity ($\chi^{(2)} \neq 0$) in particular PC and the ability for enhanced second harmonic generation due to the structural light focusing phenomenon. It is possible to realize other nonlinear conversions related to second order nonlinearity, such as, parametric conversion, low-frequency wave generation via the optical rectification and parametric decay. The latter nonlinear processes could become a basis for development of novel terahertz devices (sources, detectors, and modulators) [15–17], and these devices could be used for the the needs of terahertz spectroscopy and terahertz imaging [18-26].
Structural light focusing should be inherent to the media possessing third order nonlinearity \( (\chi^{(3)} \neq 0) \), and, thus, demonstrating the enhancement of third-order nonlinear optical processes, in particular, enhancement of third harmonic generation, is an important problem for future research. Empty 3D opal PCs made of fused silica or polymer globules [27,28] or 2D PCs based on cotton-growth ZnO nanorods [29,30] could become appropriate media for demonstrating third harmonic generation enhancement. Studying the nonlinear effect in quasi-PC structures and partially-ordered media [31-33,13] is another perspective topic for fundamental and applied research, since the structural light focusing could also appear in this structures leading to nonlinear effect enhancement, and to highly efficient interactions of light with matter.

4. Conclusion
In this paper we have reported new experimental results on enhanced second harmonic generation in PC based on the structural light focusing phenomenon. We use opal-based PC, infiltrated with NaNO\(_2\) and pumped with femtosecond laser pulses at various angles of incidence, in order to examine the dependence of second harmonic generation efficiency on the spectral position of pumping wavelength toward the PC band-gap. One order enhancement of second harmonic generation was observed in case of band-gap pumping comparing to non-band-gap pumping of the PC. In case of band-gap pumping the maximum of second harmonic generation corresponds to the direction of mirror reflection and has ±10\(^\circ\) divergence.

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References
[1] Bykov V P 1972 J. Experimental and Theoretical Physics 35 269–73
[2] Yablonovich E 1987 Phys. Rev. Lett. 58 2059–62
[3] John S 1987 Phys. Rev. Lett. 58 2486–89
[4] Meade R D, Rappe A M, Brommer K D, Joannopoulos J D, Alerhand O L 1993 Phys. Rev. B 48 8434–37
[5] Soljacic M, Joannopoulos J D 2004 Nature Materials 3 211-19
[6] Balakin A V, Bushuev V A, Mantayev B I, Ozhedev I A, Petrov E V, Shkurinov A P, Masselin P and Mouret G 2001 Phys. Rev. E 63 046609
[7] Martorell J, Vilaseca R, Corbalan R 1997 Phys. Rev. A 55 4520–25
[8] Markowicz P P, Tiryaki H, Budarah H, Prasad P N, Boyd R W 2004 Phys. Rev. Lett. 92 083903
[9] Fedyanin A A, Aktispetrov O A, Kurydyukov D A, Golubev V G, Inoue M 2005 Appl. Phys. Lett. 87 151111
[10] Zahtsev K I, Gorelik V S, Khorokhorov A M and Yurchenko S O 2014 J. Phys. Conf. Ser. 486 012003
[11] Zahtsev K I, Katyba G M, Yakovlev E V, Gorelik V S and Yurchenko S O 2014 J. Appl. Phys. 115 213505
[12] Zahtsev K I, Yurchenko S O 2014 Appl. Phys. Lett. 105 051902
[13] Yurchenko S O 2014 J. Chem. Phys. 140 134502
[14] Martienssen W, Warlimont H 2005 Springer Handbook of Condensed Matter and Materials Data (Berlin: Springer Berlin Heidelberg)
[15] Svinstov D, Vyrkov V, Yurchenko S, Otsuji T, Ryzhii V 2012 J. Appl. Phys. 111 083715
[16] Ryzhii V, Otsuji T, Ryzhii M, Leiman V G, Yurchenko S O, Mitin V, Shur M S 2012 J. Appl. Phys. 112 104507
[17] Ryzhii V, Otsuji T, Ryzhii M, Ryabova N, Yurchenko S O, Mitin V, Shur M S 2013 J. Phys. D: Appl. Phys. 46 065102
[18] Borodin A V, Panov N A, Kosareva O G, Andreeva V A, Esaulkov M N, Makarov V A, Shkurinov A P, Chin S L, Zhang X-C 2013 Opt. Lett. 38 1906–08
[19] Borodin A V, Esaulkov M N, Frolov A A, Shkurinov A P, Panchenko V Y 2014 Opt. Lett. 39 4092–95
[20] Zahtsev K I, Karasik V E, Fokina I N, Alekhnovich V I 2013 Opt. Eng. 52 068203
[21] Zahtsev K I, Gavdush A A, Karasik V E, Alekhnovich V I, Nosov P A, Lazarev V A, Reshetov I V, Yurchenko S O 2014 J. Appl. Phys. 115 193105
[22] Zahtsev K I, Chernomyrdin N V, Alekhnovich V I 2014 J. Phys. Conf. Ser. 486 012010
[23] Zahtsev K I, Gavdush A A, Lebedev S P, Yurchenko S O 2014 J. Phys. Conf. Ser. 486 012018
[24] Zaytsev K I, Kudrin K G, Koroleva S A, Fokina I N, Volodarskaya S I, Novitskaya E V, Perov A N, Karasik V E, Yurchenko S O 2014 J. Phys. Conf. Ser. 486 012014
[25] Zaytsev K I, Chernomyrdin N V, Gorevoy A V, Trofimov N E, Fokina I N, Alekhnovich V I, Karasik V E, Yurchenko S O 2014 J. Phys. Conf. Ser. 486 012034
[26] Perov A N, Zaytsev K I, Fokina I N, Karasik V E, Yakovlev E V, Yurchenko S O 2014 J. Phys. Conf. Ser. 486 012027
[27] Blanco A, Chomski E, Grabtchak S, Ibisate M, John S, Leonard S W, Lopez C, Meseguer F, Miguez H, Mondia J P, Ozin G A, Toader O, van Driel H M 2000 Nature 405 437–40
[28] Pursiainen O L J, Baumberg J J, Winkler H, Viel B, Spahn P, Ruhl T 2007 Opt. Exp. 15 9553–61
[29] Al-Hilli S M, Al-Mofarji R T, Klasen P, Willander M, Gutman N, Saar A 2008 J. Appl. Phys. 103 014302
[30] Israr M Q, Sadaf J R, Yang L L, Nur O, Willander M, Palisaitis J, Persson P O A 2009 Appl. Phys. Lett. 95 073114
[31] Vardeny Z V, Nahata A, Agrawal A 2013 Nature Photonics 7 177-87
[32] Bunkin N F, Yurchenko S O, Suyazov N V, Shkirin A V 2012 J. Biol. Phys. 38 121-52
[33] Bunkin N F, Yurchenko S O, Suyazov N V, Starosvetskiy A V, Shkirin A V, Kozlov V A 2011 Modeling the cluster structure of dissolved air nanobubbles in liquid media (Book Chapter) (New-York: Nova Science Publishers Inc.)