Enhanced third harmonic generation using the surface states of light in periodic photonic structures

Kirill I. Zaytsev1*, Vladimir S. Gorelik1,2, Gleb M. Katyba1, and Stanislav O. Yurchenko1

1 Bauman Moscow State Technical University, Moscow, Russia
2 Lebedev Physical Institute of the Russian Academy of Sciences, Moscow, Russia

*E-mail: kirzay@gmail.com

Abstract. Third harmonic generation enhancement in periodic photonic structures was experimentally observed and theoretically interpreted. Nonlinear optical effects in opal globular photonic crystals (PC) have been studied under the femtosecond laser pumping. Strong dependence of the third harmonic generation efficiency on the ratio of the central pumping wavelength and the spectral location of the PC band gaps was found. Numerical simulations based on the finite difference time-domain technique for the solution of the Maxwell’s equations were applied for investigations of the observed phenomenon origin. The simulation results have shown that the origin of the efficient nonlinear conversion is related with the surface state of electromagnetic field in PC. Interacting with the PC surface the light wavefront distorts coherently, and the effect of structure light focusing appears. Coherent wavefront distortion leads to the strong optical field localization, hence the light intensity within the certain PC regions increases. In case of the band gap pumping dramatic light redistribution appears; very sharp peaks of light intensity emerge in the region of the quartz globules, which leads to the high-efficient PC pumping.

1. Introduction and Background
An appearance of novel materials, in particular, strongly correlated optical media: periodic photonic structures, photonic crystals (PC) [1-5] and quasi-crystals [6] has attracted considerable interest in the recent time. The PC is a structure characterized by a spatially periodic distribution of the medium optical characteristics. The period of oscillations is comparable to the wavelength of light [7,8]. Periodicity of the structure leads to the dramatic changes of the photon behavior in a PC in comparison to the homogeneous medium: the spatial periodicity produces the zone structure of the dispersion relation, described within the Bloch’s effective field theory [9-10].

PCs could be used in waveguide applications [11], including PC fibers [12], for supercontinuum generation [13], for highly efficient nonlinear light conversion, high-harmonic generation and active media pumping [14-17], and for increased Raman scattering by particles injected into the PCs [18,19]. Surface states of light in PCs are an interesting phenomenon [20-27], and it could be utilized in wide range of applications, in particular, in nonlinear optics [28]. PCs are the potential basis for the creation of new optical media and devices.

The present paper is dedicated to the investigations of the third harmonic generation efficiency in opal PC, and to the research of the origin of this phenomenon by means of the numerical simulations of light interaction with the 2D PC structure.

Content from this work may be used under the terms of the Creative Commons Attribution 3.0 licence. Any further distribution of this work must maintain attribution to the author(s) and the title of the work, journal citation and DOI.

Published under licence by IOP Publishing Ltd
2. Results

2.1. Experimental observation of third harmonic generation in PC

Three samples of synthetic opal globular PC with globular diameter $a = 240.0$, 260.0 and 290.0 nm were used for experimental studying of third harmonic generation efficiency. Sample pumping was implemented utilizing the femtosecond optical pulses of Yb:KGW femtosecond laser, which worked either at the principal central wavelength ($\lambda_P = 1026.0$ nm) or at the central wavelength of the second harmonic ($\lambda_P = 513.0$ nm). The duration of the femtosecond pulse and pulse repetition rate were $\tau_p = 250.0$ nm and $f_{\text{rep}} = 200.0$ kHz, respectively. The maximal peak power of the pulse $P_{\text{max}}$ at the principle wavelength and at the wavelength of the second harmonic reached 0.7 GW and 0.35 GW, respectively. During the experiment the laser radiation of the principle mode or the second laser harmonic was focused on the surface of the sample using wide-aperture optical system with $(D/f' = 1:1)$, thus the diffraction quality of the pump spot and the peak irradiance of an object $E_{\text{max}} \approx 1.0 \text{TW/m}^2$ were achieved. High pumping intensity provided by means of ultra-short optical pulses yielded maximal efficiency of nonlinear optical conversion in PC. Electromagnetic radiation scattered by the object were detected with a diffraction grating spectrometer, which spectral sensitivity corresponds to the spectral range between 190.0 and 1000.0 nm. Spectral resolution of described system was $\Delta \lambda = 1.0$ nm.

Existence of three different PC structures with various locations of the main band gap $\lambda_{BG}$ and two pumping wavelengths $\lambda_P$ helps to study six cases of different sample pumping conditions. There are four principally different cases of the pumping wavelength $\lambda_P$ position towards the samples band gap location $\lambda_{BG}$: $1. \lambda_P = \lambda_{BG}$; $2. \lambda_P \leq \lambda_{BG}$; $3. \lambda_P \geq \lambda_{BG}$ and $4. \lambda_P \gg \lambda_{BG}$. Figure 1 shows normalized intensity of the third harmonic generated in PC for all of the cases listed above.

![Figure 1: Dependence of normalized third harmonic intensity $I_{3\omega}/I_0$ on the pumping power $P$ and the ratio between the central pumping wavelength $\lambda_P$ and the location of the PC band gap $\lambda_{BG}$.](image)

1 - $\lambda_P = \lambda_{BG}$; 2 - $\lambda_P \leq \lambda_{BG}$; 3 - $\lambda_P \geq \lambda_{BG}$; 4 - $\lambda_P \gg \lambda_{BG}$

Obviously, the condition of the effective third-harmonic generation is $\lambda_P$ matched $\lambda_{BG}$. In this case about 10.0% of the pumping intensity is transformed to the intensity of the third harmonic. Also the
efficiency of nonlinear generation in case of band gap pumping ($\lambda_P = \lambda_{BG}$) is on the order above the efficiency in case of pumping outside the band gap ($\lambda_P \gg \lambda_{BG}$).

2.2. Numerical simulation of the light interaction with the surface of PC

In order to interpret the results of the experiment (figure 1) FDTD simulation [29-31] was applied to research the structure of the surface optical modes in 2D PC. The model of the 2D PC was built of cylinders with the diameter $a = 250 \text{nm}$, which is close to the diameter of the globules of the samples researched during the experiment. The matrix of cylinders is filled with air. The refractive index of the cylinder medium is $n_1 = 2.0$, and the refractive index of the air is $n_0 = 1.0$. The size of the PC model in all directions is much bigger than the incident light wavelength $\lambda$, and a PC structure periods $T_x, T_y$: $L_x \gg \lambda, L_y \gg \lambda, L_x \gg T_x, L_y \gg T_y$. The last conditions should be satisfied to prevent non-physical distortions of the modeling results, and undesirable boundary effects. The ideally plane electromagnetic wave with wavelength $\lambda_P$ reached the PC surface from the left side.

During the simulation the time dependence of the electric field in the entire simulated space $E(x, y, t)$ was recorded; and the spatial distribution of the optical field intensity was computed the following way:

$$I(x, y) = \frac{1}{t' - t''} \int_{t'}^{t''} |E(x, y, t)|^2 dt,$$

where $c_0 = 3 \cdot 10^8 \text{ m/s}$ is a speed of light in vacuum, $t'$ and $t''$ is the time of the beginning and the end of the integration. Results of $I(x, y)$ calculations for different incident light wavelength are presented in the figure 2.

In complete accordance with the Bloch’s theory of PC [1-10,32], the observed distributions $I(x, y)$ has a periodic character in case of pumping with the wavelength $\lambda_P$ smaller than the spectral location of the first PC band gap $\lambda_{BG1}$: $\lambda_P < \lambda_{BG1}$ (figure 2a-g). In the opposite case ($\lambda_P > \lambda_{BG1}$) the structure of the optical field intensity $I(x, y)$ has homogenous character and does not contain any features (figure 2h-l). If the condition of the band gap pumping is satisfied ($\lambda_P = \lambda_{BG1} = 400.0 \text{ nm}$, $\lambda_P = \lambda_{BG2} = 200.0 \text{ nm}$) than the light intensity distribution has an average exponential decay with an increase of the sample depth $x/a$ (figure 2c and figure 2g). One could notice dramatic lateral redistribution of optical filed which could not be predicted within the usual effective field theory and associated with an effect of coherent light wavefront distortions. The distortions of the wavefront is caused by the curvature of the globule surface, and leads to the structure focusing of light in PC and appearance of the sharp peaks of light intensity concentrated in the certain regions of PC.

The sharpest peaks of intensity corresponds to the cases of band gap pumping; the intensity of light in regions of PC globules could be 10-25 times higher than the intensity of light in the equivalent homogenous optical medium in the same pumping conditions. Note, the size of the sharp light intensity peaks is smaller than the light wavelength $\lambda$, and the reason of this phenomenon is a high-aperture focusing caused by the nature of light interaction with strongly correlated optical media. Similar effects of coherent wavefront distortion and spatial light redistribution could appear in 3D PC considering the interplay between different polarization states.

Strong field localization effect observed in case of band gap pumping could be an origin of high-efficient nonlinear light conversion and enhancement of the third harmonic generation in opal PC (figure 1). To characterize the raise of the third harmonic generation efficiency the post-processing of the simulation results was implemented.
Figure 2 Spatial distribution of the electromagnetic field intensity $I(x, y)$ near the interface between the air and the 2D photonic crystal at different pumping wavelength $\lambda_P$:

(a) – $\lambda_P = 100 \text{ nm}$, (b) – $\lambda_P = 150 \text{ nm}$, (c) – $\lambda_P = 200 \text{ nm}$,
(d) – $\lambda_P = 250 \text{ nm}$, (e) – $\lambda_P = 300 \text{ nm}$, (f) – $\lambda_P = 350 \text{ nm}$,
(g) – $\lambda_P = 400 \text{ nm}$, (h) – $\lambda_P = 450 \text{ nm}$, (i) – $\lambda_P = 500 \text{ nm}$,
(j) – $\lambda_P = 550 \text{ nm}$, (k) – $\lambda_P = 600 \text{ nm}$, (l) – $\lambda_P = 650 \text{ nm}$
Figure 3 shows the results of calculation of the average light intensity cube in the volume of the PC globules \( \langle I^3 \rangle_g \) and in the volume of the PC channels filled with air \( \langle I^3 \rangle_{ch} \). The following equations were used for the calculation of this parameter dependencies on normalized pumping wavelength \( \lambda/a \):

\[
\begin{align*}
\langle I^3 \rangle_g &= \iint_{(x,y) \in V} \frac{I^3(x,y)}{I_0} \, dx \, dy, \\
\langle I^3 \rangle_{ch} &= \iint_{(x,y) \notin V} \frac{I^3(x,y)}{I_0} \, dx \, dy,
\end{align*}
\]

where \( I_0 \) is the intensity of light in the equivalent homogenous medium, for instance, in quartz. \( V \) represents the volume plurality of the PC globules.

![Figure 3](image)

**Figure 3** The dependence of the light intensity cube \( \langle I^3 \rangle \) (1) within the volume of the PC globules \( \langle I^3 \rangle_g \) and (2) within the volume of the PC channels \( \langle I^3 \rangle_{ch} \) on the normalized wavelength of \( \lambda/a \)

The intensity cube curves illustrate that the average intensity in case \( \lambda_P > \lambda_{BG1} \) (\( \lambda_P/a > 1.6 \)) is very close to the intensity in equivalent homogenous medium. In opposite case \( \lambda_P < \lambda_{BG1} \) (\( \lambda_P/a < 1.6 \)) dramatic redistribution of the light intensity appears. The intensity of quartz globule pumping raises and high efficient nonlinear conversion appears, in particular, enhanced third harmonic generation effect. One could note the peaks of \( \langle I^3 \rangle_{g(a)} \) function in the regions of first \( \lambda_{BG1} \) and second \( \lambda_{BG2} \) band gaps, which corresponds to the normalized wavelengths \( \lambda_{BG1}/a = 1.6 \) and \( \lambda_{BG2}/a = 0.8 \), respectively.

The results of numerical simulations match the experimental results of the figure 1. The efficiency of third harmonic generation raises more than ten times in case of the first band gap pumping. The nature of the effective nonlinear conversion is associated with the effect of structure light focusing in PC, the effect of dramatic redistribution of light intensity and strong localization of optical field in certain PC regions.
There are various applications of the observed optical field localization effect including enhanced nonlinear light conversion and high-efficient pumping of active media [14-17], supercontinuum generation [13] etc. Similar effects could appear in sonic (acoustic) crystals [33], because of the close analogy between electromagnetic and sound waves. The effect of structure focusing is an important one for prediction of the destructions of the quasi-sonic [33] and quasi-photonic [6,34] crystal-like constructions under the influence of acoustic and optical waves.

Observed effects are also reveal the limitations of all effective field model approaches and Bloch’s theory [1-10] to understanding periodic photonic structures, in particular, to studying strong field localizations and structure light focusing in 2D and 3D PC structures.

3. Conclusions

Third harmonic generation enhancement in PC structures was experimentally observed and theoretically interpreted. The results of the FDTD simulations of light interaction with 2D PC have shown that the origin of the efficient nonlinear conversion is associated with the surface state of electromagnetic field in PC. The structure light focusing in PC in case of band gap pumping condition is satisfied $\lambda_p = \lambda_{BG}$ leads to dramatic increase of optical field localization and to the raise of the pumping efficiency, thus the nonlinear conversion enhancement appears.

Acknowledgments

K.I.Z. and G.M.K. are grateful for the financial support of the Russian Foundation for Basic Research (RFBR), Project No. 14-02-00256. The work of V.S.G. was supported with the grants of the Russian Foundation for Basic Research (RFBR), Project No. 13-02-90420 and Project № 13-02-00449.

References

[1] Bykov V P 1972 Spontaneous emission in a periodic structure JETP. 35(2), 269 – 273
[2] Yablonovitch E 1987 Inhibited Spontaneous Emission in Solid-State Physics and Electronics Phys. Rev. Lett. 58(20), 2059 – 2062
[3] John S 1987 Phys. Rev. Lett. 58(23), 2486 – 2489
[4] Ho K M, Chan C T, Soukoulis C M 1990 Strong localization of photons in certain disordered dielectric superlattices Phys. Rev. Lett. 65(25), 3152 – 3155
[5] Yablonovitch E, Gmitter T J, Leung K M 1991 Photonic band structure: The face-centered-cubic case employing nonspherical atoms Phys. Rev. Lett. 67(17), 2295 – 2298
[6] Vardeny Z V, Nahata A, Agrawal A 2013 Optics of photonic quasicrystals Nature Photonics. 7, 177 – 187
[7] Gorelik V S 2007 Optics of globular photonic crystals Quantum Electronics. 37(5), 409 – 432
[8] Gorelik V S 2008 Optics of globular photonic crystals Laser Physics. 18(12), 1479 – 1500
[9] Meade R D, Rappe A M, Bromer K D, Joannopoulos J D, Alerhand O L 1993 Accurate theoretical analysis of photonic band-gap materials Phys. Rev. B. 48(11), 8434 – 8437
[10] Hornreich R M, Shtrikman S, Sommers C 1994 Photonic band gaps in body-centered-cubic structures Phys. Rev. B. 49(16), 10914 – 10917
[11] Vlasov Y A, O'Boyle M, Hamann H F, McNab S J 2005 Active control of slow light on a chip with photonic crystal waveguides Nature. 438, 65 – 69
[12] Knight J C, Birks T A, Russell P S J, Atkin D M 1996 All-silica single-mode optical fiber with photonic crystal cladding Optics Letters. 21(19), 1547 –1549
[13] Herrmann J, Griebner U, Zhavoronkov N, Husakou A, Nickel D, Knight J C, Wadsworth W J, Russell P S J, Korn G 2002 Experimental Evidence for Supercontinuum Generation by Fission of Higher-Order Solitons in Photonic Fibers Phys. Rev. Lett. 88(17), 173901
[14] Berger V 1998 Nonlinear Photonic Crystals Phys. Rev. Lett. 81(19), 4136 – 4139
[15] Balakin A V, Bushuev V A, Koroteev N I, Mantsyzov B I, Ozheredov I A, Shkurinov A P, Boucher D, Masselin P 1999 Enhancement of second-harmonic generation with
femtosecond laser pulses near the photonic band edge for different polarizations of incident light **Optics Letters.** 24(12), 793 – 795

[16] Balakin A V, Bushuev V A, Mantsyzov B I, Ozheredov I A, Petrov E V, Shkurinov A P, Masselin P, Mouret G 2001 Enhancement of sum frequency generation near the photonic band gap edge under the quasiphase matching conditions **Phys. Rev. E.** 63(4), 046609

[17] Soljacic M, Joannopoulos J D 2004 Enhancement of nonlinear effects using photonic crystals **Nature Materials.** 3(4), 211 – 219

[18] Rivoire K, Buckley S, Song Y 2012 Photoluminescence from In\(_{0.5}\)Ga\(_{0.5}\)As/GaP quantum dots coupled to photonic crystal cavities **Phys. Rev. B.** 85(4), 045319

[19] Gorelik V S, Zlobina L I, Sverbil P P, Fadyushin A B, Chervyakov A V 2005 Raman Scattering in Three-Dimensional Photonic Crystals **Journal of Russian Laser Research.** 26(3), 211 – 227

[20] Rosberg C R, Neshev D N, Krolikowski W, Mitchell A, Vicencio R, Molina M I, Kivshar Y S 2006 Observation of Surface Gap Solitons in Semi-Infinite Waveguide Arrays **Phys. Rev. Lett.** 97, 083901

[21] Kartashov Y V, Vysloukh V A, Torner L 2006 Surface Gap Solitons **Phys. Rev. Lett.** 96, 073901

[22] Malkova N, Ning C Z 2006 Shockley and Tamm surface states in photonic crystals **Phys. Rev. B.** 73(11), 113113

[23] Malkova N, Ning C Z 2007 Interplay between Tamm-like and Shockley-like surface states in photonic crystals **Phys. Rev. B.** 76(4), 045305

[24] Klos J 2007 Conditions of Tamm and Shockley state existence in chains of resonant cavities in a photonic crystal **Phys. Rev. B.** 76(16), 165125

[25] Ishizaki K, Noda S 2009 Manipulation of photons at the surface of three-dimensional photonic crystals **Nature** 460, 367 – 370

[26] Soboleva I V, Moskalenko V V, Fedyanin A A 2012 Giant Goos-Hänchen Effect and Fano Resonance at Photonic Crystal Surfaces **Phys. Rev. Lett.** 108(12), 123901

[27] Afshingurov B I, Bessonov V O, Nikulin A A, Fedyanin A A 2013 Observation of hybrid state of Tamm and surface plasmon-polaritons in one-dimensional photonic crystals **Appl. Phys. Lett.** 103(6), 061112

[28] Zaytsev K I, Gorelik V S, Khorokhorov A M, Yurchenko S O 2014 FDTD simulation of the electromagnetic field surface states in 2D photonic crystals **Journal of Physics: Conference Series.** 486, 012003

[29] Yee K 1966 Numerical solution of initial boundary value problems involving Maxwell's equations in isotropic media **IEEE Transactions on Antennas and Propagation.** 14(3), 302 – 307

[30] Zaytsev K I, Karasik V E, Fokina I N, Alekhnovich V I 2013 Invariant embedding technique for medium permittivity profile reconstruction using terahertz time-domain spectroscopy **Optical Engineering.** 52(6), 068203

[31] Fokina I., Zaytsev K I, Karasik V E, Tsapenko K I 2013 Scattering of terahertz radiation in thin layers of dielectric materials **Proceedings of SPIE.** 8846, 88460A-1

[32] Sipe J E, Bhat N A R, Chak P, Pereira S 2004 Effective field theory for the nonlinear optical properties of photonic crystals **Phys. Rev. E.** 69, 016604

[33] Liu Z, Zhang X, Mao Y, Zhu Y Y, Yang Z, Chan C T, Sheng P 2000 Locally Resonant Sonic Materials **Science.** 289(5485), 1734-1736

[34] Bunkin N F, Yurchenko S O, Suyazov N V, Shkirin A V 2012 Structure of the nanobubble clusters of dissolved air in liquid media **Journal of Biological Physics.** 38, 121-152