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Frequency doubling in periodic nonlinear photonic crystals mediated by random layers

Andrey M. Vyunishev[1,2,*] and Anatoly S. Chirkin[3]

[1]Kirensky Institute of Physics, Federal Research Center KSC SB RAS, Krasnoyarsk 660036, Russia
[2]Department of Photonics and Laser Technology, Siberian Federal University, Krasnoyarsk 660079, Russia
[3]Faculty of Physics and International Laser Center, M. V. Lomonosov Moscow State University, Moscow 119992, Russia
*Corresponding author: vyunishev@iph.krasn.ru

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We study the frequency doubling in a quadratic nonlinear photonic crystal consisting of periodically poled structures mediated by uniform layers with random lengths. These structures can be formed by new local impact methods for ferroelectric crystal structuring. The statistical frequency doubling theory is developed for such structures. The effect of the number of random layers and variation in their thicknesses on the second-harmonic conversion efficiency is clarified. It is demonstrated that a proper choice of the intermediate layer thickness can enhance or suppress the conversion efficiency. A new type of the Maker-fringes-like second-harmonic intensity oscillations is predicted.

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At present, nonlinear photonic crystals (NPCs) are widely used for conversion of coherent laser radiation into new frequencies [1,2]. Nonlinear photonic crystals combine the high conversion efficiency, compactness, and exceptional universality in designing structures for target nonlinear optical processes. For these purposes, electric field poling of ferroelectric crystals was developed [3]. At the same time, local impact methods for structuring ferroelectrics have recently been proposed to fabricate NPCs. These methods include electron beam writing [4,5], atomic force microscope writing [6,7], and direct laser beam pattering [8,9]. The advantage of these methods is the possibility of fabricating arbitrary high-quality domain structures on submicrometer and nanosized scales [10]. The drawback of these methods is the restriction imposed on the writing area size, which typically varies within 0.02–0.2 cm. Fabrication of relatively large (about 1 cm and more) structures faces certain difficulties. Although these structures can gradually be formed, the unavoidable random phase shifts at the transition between specific regular sections are caused by insufficiently accurate sample positioning during fabrication, which can result in the low nonlinear conversion efficiency.

The statistical phenomena related to the randomness of the nonlinear media parameters drew attention to nonlinear optics long time ago (see studies [11,12] and monograph [13] and references therein). Random spatial distribution of nonlinearity leads to phase errors, which disturb phase matching [14]. On the other hand, it suggests random quasi-phase matching, which enables widely tunable [15,16] and broadband frequency conversion [17]. The earlier studies addressed either the continuously distributed random variations in domain thicknesses along the structures [18–22], or random fluctuations of the domain wall positions in regular structures [23].

In view of the aforesaid, the effect of random phase shifts on the nonlinear optical processes in NPCs fabricated by the new local impact methods is of special interest. In this work, we investigate second-harmonic generation (SHG) in an assembled nonlinear photonic crystal with periodically inserted random layers and establish a new type of the second-harmonic (SH) intensity oscillations.

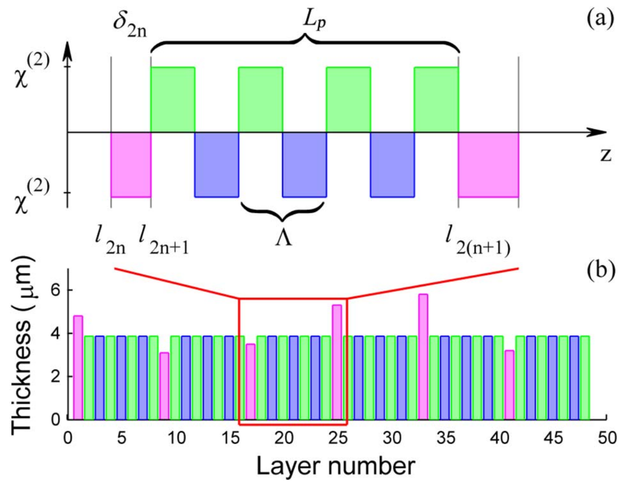

Fig. 1. (a) Nonlinearity modulation in the assembled structure composed of alternating periodic layers (blue and green) and random uniform layers (magenta). (b) Layer thicknesses versus layer numbers in the structure.

We consider the NPC design shown in Fig. 1. The NPC structure stacked in the direction of the z-axis is assembled from periodically poled structures mediated by uniform layers with the thickness fluctuations. The average random layer thickness was taken to be equal to a half-period $\Lambda/2$ of the periodic structure and the layer thickness fluctuations were assumed to obey the Gaussian distribution law. For certainty, we assume the number of random layers to be equal to the number $N+1$ of the periodically poled structures consisting of $2M+1$ domains. In the undepleted fundamental wave approximation, the SH amplitude is governed by

$$A_2 = -i\beta A_1^2 F(L), \tag{1}$$

where $\beta = 4\pi^2\chi^{(2)}/\lambda_2 n(\lambda_2)$ is the nonlinear coupling coefficient, A_1 is the fundamental frequency (FF) amplitude, λ_2 is the SH wavelength, $n(\lambda_2)$ is the SH refractive index, $\chi^{(2)}$ is the quadratic nonlinear susceptibility, and L is the crystal's length. The function $F(L)$ describes the contribution of the structure to the process and takes the simplest form $F(L) = L$ under the phase-matching conditions.

According to Fig. 1, the SH amplitude is determined by the superposition of the amplitudes generated by the uniform (u) and periodically poled (p) layers:

$$F(L) = \sum_{n=0}^{N} [F_u(2n, 2n+1; \delta_{2n}) + F_p(2n, 2(n+1); L_p)]. \tag{2}$$

Here, the functions F_u and F_p correspond to the uniform and periodically poled structure parts, which are

$$F_u(2n, 2n+1; \delta_{2n}) = \frac{1}{i\Delta k}(e^{i\Delta k l_{2n+1}} - e^{i\Delta k l_{2n}}), \tag{3}$$

$$F_p(2n+1, 2(n+1); L_p) = \int_{l_{2n+1}}^{l_{2(n+1)}} g(z) e^{i\Delta k z} \mathrm{d}z. \tag{4}$$

In Eqs. (3) and (4), l_n is the layer coordinate ($l_0 = 0$), $\Delta k = k_2 - 2k_1$ is the wave vector mismatch, $k_{1,2}$ are FF and SH wave vectors directed along the z-axis, $g(z)$ is the alternating periodic function of the coordinate z, $\delta_{2n} = l_{2n+1} - l_{2n}$ is the uniform layer thickness (random variable), $L_p = l_{2(n+1)} - l_{2n+1} = \frac{1}{2}(2M+1)\Lambda$ is the periodic part thickness, and M is the number of periods. It can be shown that $l_{2n} = \sum_{q=0}^{n-1} \delta_{2q} + nL_p$, where the first term summarizes random thicknesses of uniform layers.

Equation (4) can be transformed to

$$F_p(2n+1, 2(n+1); L_p) = -i\frac{2}{\pi} L_p e^{i\Delta k l_{2n+1}}, \tag{5}$$

where $\Delta k l_{2n+1}$ is the phase between the SH wave and polarization induced at the double frequency.

Taking into account Eqs. (3) and (5), we arrive at

$$F = -i\sum_{n=0}^{N}\left[\left(\frac{2L_p}{\pi} + \frac{1}{\Delta k}\right) e^{i\Delta k l_{2n+1}} - \frac{1}{\Delta k} e^{i\Delta k l_{2n}}\right]. \tag{6}$$

Further, with the accuracy $\mu = \pi/(2|\Delta k|L_p) = \Lambda/4L_p \ll 1$ (according to the parameters of the structure specified below $\mu \approx 10^{-2}$), we use Eq. (6) in the form

$$F \approx -i\sum_{n=0}^{N}\left[\left(\frac{2L_p}{\pi}\right) e^{i\Delta k l_{2n+1}}\right]. \tag{7}$$

Now we substitute Eq. (7) in Eq. (1) and multiply A_2 by its complex conjugate, designating at the same time summing indices as n_1 and n_2, respectively. As a result, the SH intensity is

$$I_2 = |A_2(L)|^2 = \beta^2 I_1^2 \left(\frac{2L_p}{\pi}\right)^2 \sum_{n_1,n_2=0}^{N} [e^{i\Delta k(l_{2n_1+1} - l_{2n_2+1})}], \tag{8}$$

where

$$\Delta k(l_{2n_1+1} - l_{2n_2+1}) = \Delta k\left[(n_1 - n_2)L_p + \sum_{q=n_2}^{n_1} \delta_{2q}\right]. \tag{9}$$

Assume the uniform layer thickness to be

$$\delta_{2q} = (1 + \sigma\xi_{2q})\Lambda/2, \tag{10}$$

where $\sigma\xi_{2q}$ specifies the Gaussian fluctuations of uniform layer thicknesses, σ is the dispersion, ξ_{2q} is the random quantity ($\langle\xi_{2q}\rangle = 0$, $\langle\xi_{2q}^2\rangle = 1$) with the fluctuations for different q being statistically independent. Statistical averaging of Eq. (8) over a set of implementations of the structure, according to Eqs. (10) and (9), yields

$$\langle I_2\rangle = \beta^2 I_1^2 \left(\frac{2}{\pi}L_p\right)^2 \sum_{n_1,n_2=0}^{N,N} \exp\left(-\frac{(\pi\sigma)^2}{2}|n_1 - n_2|\right). \tag{11}$$

If $(\pi\sigma)^2 \gg 1$, then the summation of the SH amplitudes will be incoherent and we obtain

$$\langle I_2^{nch}\rangle = \beta^2 I_1^2 \left(\frac{2}{\pi}L_p\right)^2 (N+1). \tag{12}$$

At the coherent addition of the SH amplitudes ($\sigma = 0$), we have

$$I_2^{ch} = \beta^2 I_1^2 \left(\frac{2}{\pi}L_p\right)^2 (N+1)^2. \tag{13}$$

After double summation in Eq. (11), we obtain the following general result:

$$\begin{aligned}\langle I_2\rangle = {} & \beta^2 I_1^2 \left(\frac{2}{\pi}L_p\right)^2 (1 - e^{-\gamma})^{-2} \\ & \times [2(N+1) - 2(N+2)e^{-\gamma} \\ & + 2e^{-\gamma(N+2)} - (N+1)(1 - e^{-\gamma})^2].\end{aligned} \tag{14}$$

Here, $2\gamma = (\pi\sigma)^2$ is the phase dispersion. Using L'Hospital's rule twice, in the limit $\gamma \to 0$, Eq. (14) is reduced to Eq. (13), while at $\gamma \gg 1$ we obtain Eq. (12). The results of single random implementations are intermediate between these two cases.

We consider frequency doubling of Nd:YAG laser radiation at a wavelength of 1.064 μm. The lithium niobate crystal was chosen as a nonlinear medium with the refractive index dispersion from [24]. The wave vector mismatch for the *e-ee* interaction is $\Delta k \approx 0.92\ \mu\mathrm{m}^{-1}$. Thus, the first-order quasi-phase-matched SHG requires modulation of the nonlinearity with a period of $\Lambda = 6.78\ \mu\mathrm{m}$.

A set of random uniform layer thicknesses is simulated using Eq. (10). In practice, it is reasonable to take into account the absolute accuracy of sample positioning $\sigma\Lambda/2$ instead of the relative σ value. As an actual positioning accuracy, we can choose a value of 1 μm corresponding to the relative standard deviation $\sigma \approx 0.15$.

Second-harmonic intensity $I_2 = |A_2(L)|^2$ is calculated using Eqs. (1) and (6). Dependences of SH intensity on the coordinate l_n for a series of random implementations of the structure are shown in Fig. 2 (blue curves, 1–3). Random implementations of the structure were simulated using Eq. (10). The parameters were $\sigma = 0.15$, $M = 30$, $N = 24$, and a crystal thickness of $L \approx 5$ mm. We found that the monotonic SH intensity growth in the periodically poled layers can be changed for a decrease due to the random phase shifts acquired in the random uniform layers, and then the process can be switched back (curves 2 and 3). The average SH intensity calculated by Eq. (14) is raised almost linearly at N (red curve in Fig. 2).

Figure 3 shows the SH intensity as a function of the standard layer thickness deviation for 10 random implementations calculated using Eqs. (6) and (10). The data averaging reveals the nonmonotonic behavior, which is caused by a small number of implementations. One can see that the average SH intensity described by Eq. (14) derived using the developed statistical theory fits the results of the random implementations well.

We consider the process in the extreme cases when the uniform layer thickness is exactly equal to the regular structure half-period or period. The first case corresponding to the perfect quasi-phase matching is mathematically expressed as $\delta_n = \Lambda/2$ and $\sigma = 0$. In this case, the SH intensity calculated using Eq. (12) obeys the quadratic law corresponding to the quasi-phase-matched SHG (Fig. 2).

An interesting situation is observed when the uniform layer thicknesses are exactly equal to the regular structure period ($\delta_n = \Lambda$ and $\sigma = 0$). Basically, the SHG phase mismatch leads to the SH intensity oscillations along the media. In the case under study, a new type of the SH intensity oscillations along the structure is numerically predicted, as can be seen in Fig. 4. From our model it is possible to find that extreme values of the SH intensity are given by dependence $I_2 \sim \sin^2(\Delta k N \tilde{\Lambda}/4)$,

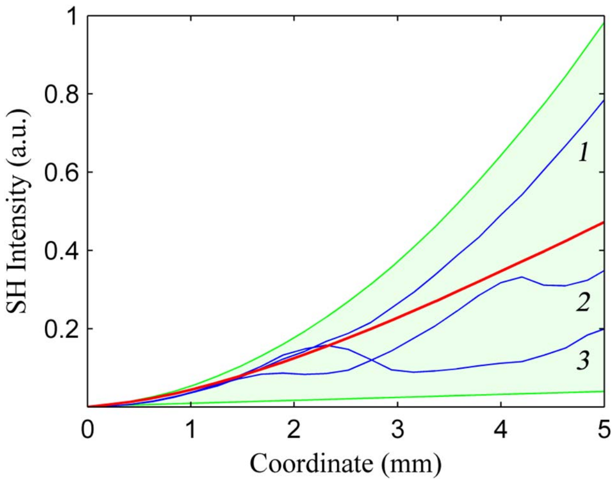

Fig. 2. Dependences of the SH intensity on the coordinate (blue curves, 1–3) for three random implementations of the structure calculated using Eq. (8) ($\sigma = 0.15$, $M = 30$, $N = 24$, and $L \approx 5$ mm), and average intensity calculated using Eq. (14) (red curve). The outlined region is constrained by the results calculated using Eq. (12) (lower green curve) and Eq. (13) (upper green curve).

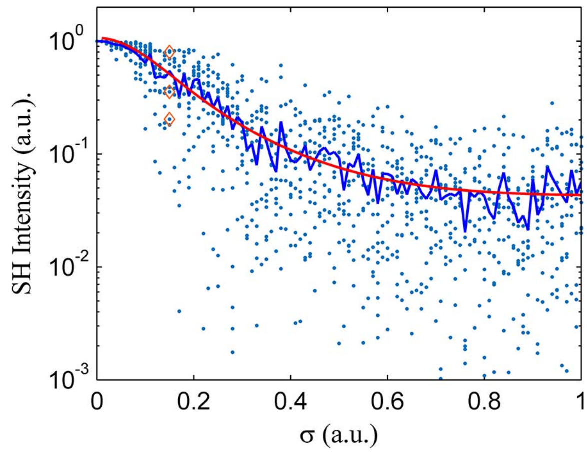

Fig. 3. Dependence of the SH intensity on the standard random layer deviation for 10 random implementations (blue points), their average values (blue nonmonotonic curve), and average intensity calculated using Eq. (14) (red monotonic curve).

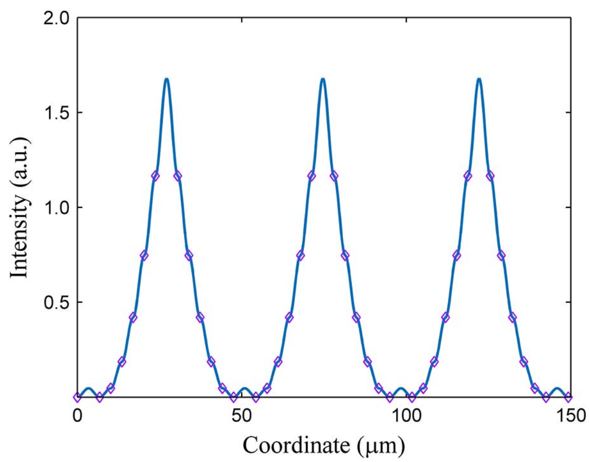

Fig. 4. SH intensity oscillations in the structure calculated numerically (solid line) and using Eq. (6) (points) at the parameters of $\delta_n = \Lambda$, $\sigma = 0$, and $M = 2$.

where $\tilde{\Lambda} = (2M + 3)\Lambda$ is the oscillation period. It differs from the SH intensity oscillations with period Λ in uniform media. In the case under study the phase shift caused by the wave vector mismatch is defined by a half of the length of the uniform layer and is equal to π. The phase shift is equal to $m\pi$ for the structure with m periods of the structure. It results in periodic dependence of intensity with the period equal to the double length of the periodically poled layer. The oscillations are very similar to those corresponding to the Maker fringes observed in uniform media at the phase mismatch (see, for example, [22]). The oscillation amplitude can be enhanced by a factor of $4(M + 1)^2$. In addition, this behavior will result in the SH intensity oscillations in the spectral and angular dependences shown in Fig. 5(a). They can be treated as quasi-phase-matched Maker fringes. For this purpose, the fundamental wavelength was tuned and thicknesses of all the layers were divided by $\cos\ \theta$, where angle θ is counted from the quasi-phase-matching

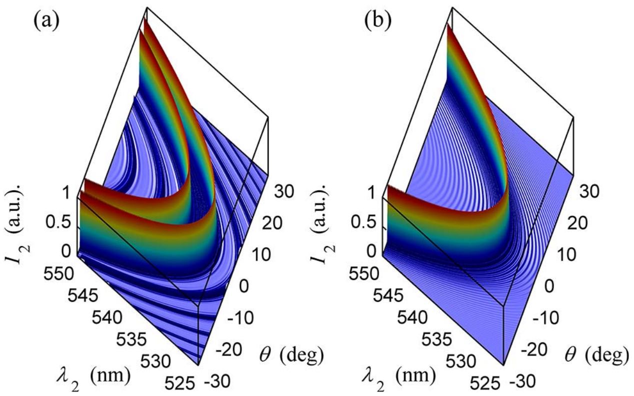

Fig. 5. Normalized SH spectral intensity versus NPC rotation angle ($M = 30$ and $N = 7$): (a) $\delta_n = \Lambda$ and (b) $\delta_n = \Lambda/2$.

direction. There are two spectral peaks that exhibit a long-wavelength shift upon rotation of the structure. The spectral spacing between the peaks grows with a decrease in M. Figure 5(b) shows the corresponding angular dependence of the SH spectral intensity for a periodically poled crystal ($\delta_n = \Lambda/2$).

Recently we have known that similar structures with random [25] and regular [26] intermediate layers were previously studied. Our study complements these results, which primarily focused on spectral features of the second-harmonic. At the same time, we have accurately developed a statistical approach which can be generalized to the study of frequency doubling of ultrashort laser pulses in such structures.

Thus, we have developed the statistical theory of frequency doubling in assembled nonlinear photonic crystals with the periodic structures mediated by the layers of random thickness. Our analysis shows that the SH intensity in the structure under study in the limit incoherent case is $I_2 \propto NM^2$ instead of $I_2 \propto (NM)^2$ corresponding to the regular structure (coherent summation of the contribution of layers). It means that the structure under study is inferior to the regular one in N times. It was shown that the second-harmonic oscillations enhanced by the quasi-phase matching take place in the absence of periodic compensations of the phase mismatch.

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