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Quasi-phase-matched second-harmonic generation in nonlinear crystals with harmonic modulation of nonlinearity

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Abstract

We propose a new type of nonlinear photonic crystal with the lattice representing a harmonic function of the propagation coordinate. This type of spatial modulation of the quadratic nonlinearity is implemented to enhance the second-harmonic generation (SHG) efficiency. The process resembles that realized in periodic nonlinear photonic crystals with rectangular nonlinearity modulation under quasi-phase-matching. Residual periodic oscillations of the second-harmonic intensity were found. Comparison of the unipolar and bipolar modulations showed that they are equivalent with regard to the oscillations. The SHG efficiency in the harmonic lattices is 1.6 times lower compared with the efficiency corresponding to the quasi-phase-matching in periodic nonlinear photonic crystals with rectangular lattices.

Keywords: second-harmonic generation, quasi-phase-matching, nonlinear photonic crystals

(Some figures may appear in colour only in the online journal)

1. Introduction

At the dawn of nonlinear optics, it was established that high-efficiency nonlinear frequency conversion in a transparent nonlinear medium requires the phase-matching condition to be met. The physical meaning of this condition is the conservation of full momentum of interacting waves. In general, this can be obtained by using angular phase-matching in birefringent nonlinear media [1] or quasi-phase-matching in artificially structured nonlinear media [2–5], also known as nonlinear photonic crystals (NPCs) [6]. The latter approach, which was proposed earlier, uses spatial structuring of media for compensating the phase mismatch. The conventional technique for fabricating NPCs is based on the electric field poling of ferroelectric crystals [7], which implies the rectangular spatial modulation of nonlinearity (bipolar rectangular lattice). As a result, the phase mismatch is periodically compensated each time the phase difference of interacting waves attains π. This means that the rate of conversion to the harmonic wave is not optimal and varies within a structure period. It limits the total conversion efficiency relative to the case of angular phase matching. At the same time, NPCs demonstrate wide capabilities for implementing nonlinear optical processes via choosing a proper structure period. In such structures, the choice of polarizations of interacting waves is not restricted, and, therefore, the maximum nonlinear coefficients, like d_{33} in lithium niobate, can be employed. Until now, different NPC designs have been studied, which offer a binary spatial modulation of nonlinearity. Among them are deterministic structures, including Fibonacci [8], Thue–Morse [9], superimposed [10, 11], and random structures [12–14]. Recently, a new method for structuring the materials has been developed [15], which allows continuous nonlinearity distribution over the structure in accordance with a specified law. Therefore, it is of interest to consider new types of nonlinearity modulation that would yield the optimal conversion efficiency and combine the most advantages of uniformly poled and structured crystals. At the same time, much attention is focused upon the harmonic modulation of the physical quantity, which has been an object of numerous studies in different fields of physics, including atomic, solid-state and wave phenomena physics, acousto-optics etc. An exception is nonlinear frequency conversion, in which the harmonic modulation has not been employed yet. Recently, the first realizations of three-dimensional (3D) NPC structures in a core of lithium niobate waveguide were reported [16]. Among these are helical twisted structures that convert a fundamental Gaussian beam into a vortex second-harmonic beam. This approach [16] can be used for structuring NPCs with smooth, harmonic or arbitrary modulation of nonlinearity that represents a particular interest for conversion of radiation with the specific characteristics.

In this paper, we consider for the first time the second-harmonic generation (SHG) in an NPC of the new type, the spatial lattice of which is a harmonic function of the propagation coordinate. In addition, possible experimental implementations of the arbitrarily structured NPCs are discussed.

2. Theoretical analysis and results

In the most general case, the harmonic spatial modulation of the quadratic nonlinear coefficient can be written as

$$d(z) = d_{\mathrm{eff}}\left(a + f\sin Gz\right). \tag{1}$$

Here, d_{eff} is the effective nonlinear coefficient, f is the amplitude of the nonlinearity modulation as compared with the average amplitude a ($|a| + |f| \leqslant 1$), $G = 2\pi/\Lambda$ is the primary reciprocal lattice vector (RLV), and Λ is the structure period. Note that $|a| \geqslant f$ corresponds to unipolar modulation (UPM) and $|a| < f$ corresponds to bipolar modulation (BPM). We will refer to the balanced modulation of nonlinearity if the average value is $a = 0$. Examples of the unipolar and bipolar functions of the nonlinear coefficient modulation calculated using (1) are shown in figure 1.

In the undepleted fundamental amplitude approximation, the SHG process in a medium with an arbitrarily varying nonlinearity $d(z)$ is governed by equation [1]:

$$A_2 = \frac{4\pi k_2}{n_2}A_1^2\int d(z)\mathrm{e}^{-\mathrm{i}\Delta kz}dz, \tag{2}$$

where A_1 is the fundamental frequency amplitude, $\Delta k = k_2 - 2k_1$ is the wavevector mismatch, and n_2 is the second-harmonic (SH) refractive index. Substituting the structure described by (1) into the integral in (2), we obtain

$$A_2 = a\Gamma\int \mathrm{e}^{-\mathrm{i}\Delta kz}dz + f\Gamma\int \sin Gz\mathrm{e}^{-\mathrm{i}\Delta kz}dz, \tag{3}$$

where $\Gamma = 4\pi k_2 d_{\mathrm{eff}}A_1^2/n_2$ is the nonlinear coupling coefficient. Integrating (3) over $[0, L]$, we arrive at the following expression:

$$\begin{aligned} A_2 = &-\frac{a\Gamma}{\mathrm{i}\Delta k}\left(\mathrm{e}^{-\mathrm{i}\Delta kL} - 1\right) \\ &+\frac{f\Gamma}{G^2 - \Delta k^2}\left[G - \mathrm{e}^{-\mathrm{i}\Delta kL}\left(G\cos GL + \mathrm{i}\Delta k\sin GL\right)\right]. \end{aligned} \tag{4}$$

The dependence of SH intensity obtained using (4) ($I_2 = |A_2|^2$) on the effective coordinate z/Λ is presented in figure 2 for a uniformly poled nonlinear crystal (UPC) and for the structures shown in figure 1. Refractive indexes of $n_1 = 1.450$ and $n_2 = 1.460$ are taken for fused silica [17] at a fundamental frequency wavelength of 1064 nm and for the SH, respectively. To avoid uncertainty in the calculations using (4), a small wavevector mismatch was introduced ($G - \Delta k = 10^{-5}$). The first term in (4) ($f = 0$) represents the field amplitude generated in the UPC with nonzero nonlinearity $a = 0.5$ resulting in weak-intensity oscillations. The second term corresponding to the BPM ($a = 0, f = 0.5$) shows the nearly quadratic spatial dependence of the intensity modulated at the spatial frequency Δk. Taking into account both terms in (4) under the condition $f < |a|$, we arrive at the case of UPM and obtain the more complex spatial dependence of the SH intensity with large-scale spatial oscillations. The respective spatial frequency is twice as low as in the case of BPM, $G' = \Delta k/2$. The scale of these oscillations depends on the fraction f/a. The lower the fraction f/a, the weaker the oscillations are. In the limit $f \ll |a|$, we obtain the case of non-phase-matched SHG in UPC and the regular SH-intensity oscillations are still observed (figure 2). Therefore, the amplitude of nonlinearity f makes the largest contribution to the SH intensity yield, while the average value a strengthens the intensity of oscillations. It can be seen in figure 2 that the unipolar and bipolar modulations are equivalent with regard to the oscillations. Note that the balanced BPM ($a = 0$) covers the full modulation range ($2f$ in (1)) resulting in a fourfold SH intensity gain. Hence, without loss of generality, we may consider the balanced BPM to be described as

$$d(z) = d_{\mathrm{eff}}f\sin Gz. \tag{5}$$

In this case, (4) reads

$$A_2 = \frac{f\Gamma}{G^2 - \Delta k^2}\left[G - \mathrm{e}^{-\mathrm{i}\Delta kL}\left(G\cos GL + \mathrm{i}\Delta k\sin GL\right)\right]. \tag{6}$$

Alternatively, (6) can be presented in the form

$$\begin{aligned} A_2 = \frac{f\Gamma G}{G^2 - \Delta k^2}\Bigg[1 - \frac{1}{2}\left(1 + \frac{\Delta k}{G}\right)\mathrm{e}^{-\mathrm{i}(\Delta k - G)L} - \\ \frac{1}{2}\left(1 - \frac{\Delta k}{G}\right)\mathrm{e}^{-\mathrm{i}(\Delta k + G)L}\Bigg]. \end{aligned} \tag{7}$$

If $G \approx \Delta k$, the third term can be omitted and (7) is reduced to the well-known form

$$A_2 = -\frac{f\mathrm{i}\Gamma L}{2}\mathrm{e}^{-\mathrm{i}(\Delta k - G)L/2}\,\mathrm{sinc}[(\Delta k - G)L/2]. \tag{8}$$

Here, $\mathrm{sinc}(x) \equiv \sin(x)/x$. If $G - \Delta k \approx 0$, the SH intensity obeys the quadratic law, as shown in figure 2.

Figure 3 shows the dependence of the SH intensity on the effective coordinate calculated using (6) ($I_2 = |A_2|^2$). In this case, a significant increase in the SH intensity is observed. The SH intensity is nearly a quadratic function of the coordinate, but the weak-residual-intensity oscillations still remain. A 1.6 times decrease in the SHG efficiency is observed in the harmonic NPC relative to the angular phase-matching in the UPC. However, the use of the maximal nonlinear coefficients would yield a higher conversion efficiency in the harmonic NPCs than in the uniformly poled crystals.

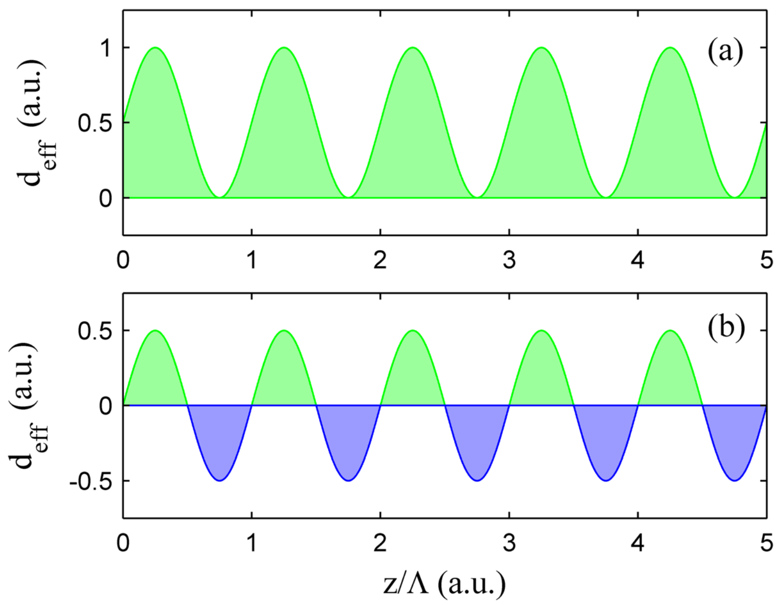

Figure 1. Examples of unipolar (a) and bipolar (b) modulation of nonlinear coefficient ($f = 0.5$).

It is especially interesting to compare the SH yield in the harmonic and rectangular structures with similar parameters. The rectangular modulation of the nonlinear coefficient is described by the formula

$$d_r(z) = d_{\text{eff}}\,(a + f\,\text{sgn}[\sin Gz])\,. \tag{9}$$

Here, $\text{sgn}(x) \equiv x/|x|$ is a signum function. The spatially varying part of (9) can be presented as a Fourier series, i.e.

$$d_r(z) = d_{\text{eff}}\left(a + f\sum_m g_m \mathrm{e}^{-\mathrm{i}mG(z+\Lambda/4)}\right), \tag{10}$$

where g_m are the Fourier coefficients, such that $g_m = 0$ at the even m values and $g_m = (2/\pi)\,m$ at the odd m values. In (10), the term $\Lambda/4$ is introduced to account for the spatial shift of the rectangular lattice to adjust the maxima with those of the harmonic one. Thus, substituting (10) into (2), we obtain

$$\begin{aligned} A_2 = &-\frac{a\Gamma}{\mathrm{i}\Delta k}\left(\mathrm{e}^{-\mathrm{i}\Delta kL} - 1\right) \\ &+ f\Gamma L\sum_m g_m \mathrm{e}^{-\mathrm{i}\left(\Delta k' + \Lambda/2\right)L/2}\,\text{sinc}\left(\Delta k' L/2\right), \end{aligned} \tag{11}$$

where $\Delta k' = \Delta k - mG$.

We limit the consideration to the case of a rectangular BPM, i.e. $a = 0$ in (9) and (11) ($m \in [-30, 30]$). The results of the calculations are presented in figure 3. It can be seen that the SH intensity grows faster than the SH intensity corresponding to the harmonic nonlinearity modulation. Comparison of (8) and (11) ($a = 0$) showed that the SH conversion efficiency in the rectangular structure increased by a factor of $(G_m/2)^2$, which is $16/\pi^2 \approx 1.6$ for the first-order QPM. Thus, the rectangular nonlinearity modulation is optimal for the quasi-phase-matched SHG and any changes in the modulation type result in a conversion efficiency drop. Moreover, the term 'QPM-order' for a harmonic structure becomes insignificant, because an increase in the SH conversion efficiency is only possible when the phase mismatch is fully compensated by the primary

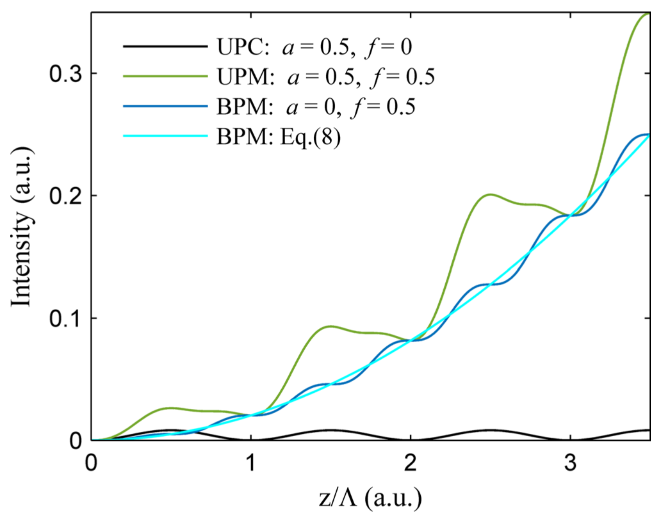

Figure 2. SH intensity ($I_2 = |A_2|^2$) versus coordinate for uniformly poled crystals ($f = 0$) and for nonlinear crystals with harmonic modulation of the nonlinear coupling coefficient.

RLV, i.e. at $\Delta k = G$ instead of $\Delta k = mG$, which corresponds to the rectangular structure. In addition, comparison of (8) and (11) shows that the spectral bandwidth corresponds to that of the periodic structure with the rectangular nonlinearity modulation [3] and takes the form

$$\Delta\lambda \simeq \frac{4}{L}\sqrt{\frac{\ln 2}{\gamma}}\left|\frac{d\Delta k}{d\lambda}\right|^{-1}, \tag{12}$$

where $\gamma \approx 0.36$, and λ is the fundamental wavelength.

The structure described by (1) can be fabricated on the basis of porous quartz filled with sodium nitride [15]. The nonlinear coefficient is modulated by varying the volume fraction of the nonlinear material in porous quartz (filling factor). It is noteworthy that the sub-micrometer spatial periodicity can be obtained by this method. This approach greatly extends the range of materials suitable for efficient frequency conversion,

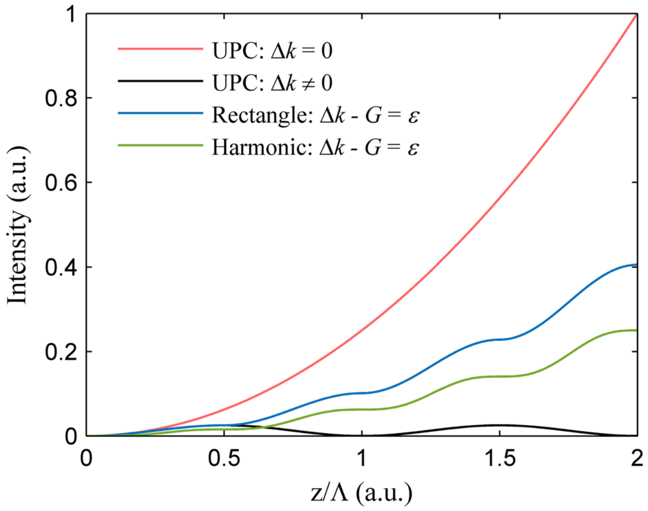

Figure 3. SH intensity versus effective coordinate in the uniformly poled crystals and the rectangular and harmonic BPMs ($\varepsilon = 10^{-5}$).

including non-ferroelectric crystals and low-birefringent materials. In particular, using this method, the efficiency of conversion to ultraviolet and vacuum ultraviolet [18–20] can be enhanced. The unipolar rectangular nonlinearity modulation can be performed using the 60- or 90-degree domain structures in ferroelectrics or poled glasses.

3. Conclusion

We studied SHG in media offering a new type of harmonic spatial nonlinearity modulation. This type of spatial modulation is implemented to increase the conversion efficiency via the periodic compensation of phase mismatch. Periodic oscillations of the SH intensity in a unipolar harmonic lattice were revealed. The unipolar and bipolar modulations were found to be equivalent with regard to the oscillations. A fourfold decrease in the SHG efficiency in harmonic lattices compared with the efficiency corresponding to the phase-matched SHG in uniformly poled crystals was found. Nevertheless, the conversion efficiency typical of uniformly poled crystals can be attained in the investigated crystals using higher nonlinear coefficients. The harmonic nonlinearity modulation can be used to implement different parametrical interactions in a wide application range, including vacuum ultraviolet generation.

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