Phase matching in nonlinear $\chi^{(2)}$ photonic crystals

Solomon Saltiel

Faculty of Physics, University of Sofia, 5 J. Bourchier Boulevard, 1164 Sofia, Bulgaria

Yuri S. Kivshar

Optical Sciences Center, Research School of Physical Sciences and Engineering, Australian National University, Canberra, ACT 0200, Australia

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We analyze harmonic generation in a two-dimensional (2D) $\chi^{(2)}$ photonic crystal and demonstrate the possibility of multiple phase matching and multicolor parametric frequency conversion. We suggest a new type of photonic structure to achieve simultaneous generation of several harmonics; we also present both general analytical results and design parameters for 2D periodically poled LiNbO₃ structures. © 2000 Optical Society of America

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Photonic crystals are usually viewed as optical analogs of semiconductors that modify the properties of light similarly to a microscopic atomic lattice that creates a semiconductor bandgap for electrons.¹ It is therefore believed that by replacing relatively slow electrons with photons as the carriers of information one can dramatically increase the speed and the bandwidth of advanced communication systems. Recent fabrication of photonic crystals with a stop band at optical wavelengths from 1.35 to 1.95 μ m (Ref. 2) makes this promise realistic.³

Nonlinear photonic crystals with quadratic [or $\chi^{(2)}$] periodic nonlinear susceptibility can be effectively employed for parametric frequency conversion and second-harmonic generation (SHG), as was first suggested theoretically⁴ and recently verified experimentally for a two-dimensional (2D) LiNbO3 photonic structure.5 The authors of Ref. 5 also observed third- and fourth-harmonic generation in a single $\chi^{(2)}$ structure, which are usually associated with cascaded second-order processes but are difficult to distinguish experimentally.⁶ In this Letter we demonstrate theoretically that 2D $\chi^{(2)}$ photonic crystals permit simultaneous phase matching of several parametric processes and thus provide an ideal environment for the experimental realization of different types of multistep cascading effect theoretically predicted earlier.⁷ We also present the design parameters for observing multiple harmonic generation in 2D periodically poled LiNbO3 structures of lower symmetry.

First, we consider a 2D $\chi^{(2)}$ photonic crystal with a symmetric triangular lattice^{4,5} for which the linear susceptibility is constant in the whole material but the sign of the second-order susceptibility $\chi^{(2)}$ varies periodically, as shown in Fig. 1. The triangular lattice is characterized by only one parameter, the lattice spacing d. We call this type of 2D grating a symmetric nonlinear photonic crystal (SNPC). As can easily be seen from Fig. 1, this 2D structure creates an infinite number of rows (directions of grating) along which the sign of the second-order nonlinearity varies periodically with different periods. Such gratings can be used as one-dimensional periodic structures for quasi-phase matching (QPM) of the nonlinearity-induced harmonic generation, as usually achieved in one-dimensional structures based on periodically poled $LiNbO_3$ or $LiTaO_3$.⁸

Each of the effective gratings in a SNPC (with the indices j = a, b, c, ...) is characterized by period Λ_j and the direction of the equivalent wave vector K_j . For example, for grating a in Fig. 1, we have $\Lambda_a = (\sqrt{3}/2)d$ and $K_a = (4\pi/d\sqrt{3})$. To find possible phase-matching processes for a 2D crystal, we should use the reciprocal lattice⁹ formed by the fundamental vectors of two gratings, \mathbf{K}_a and \mathbf{K}_c , for which $|\mathbf{K}_a| = |\mathbf{K}_c|$ [see Fig. 2(a)]. All other grating vectors can be found as $\mathbf{K}_{pq} = p\mathbf{K}_c + q\mathbf{K}_a$, where p and q are integer (positive or negative) numbers. For the geometry shown in Fig. 1, we find that $\mathbf{K}_a = (0, K_a)$, $\mathbf{K}_c = (K_0\sqrt{3}/2, K_0/2)$, and $|K_{qp}| = K_0(p^2 + q^2 + pq)^{1/2}$, where $K_0 = 4\pi/d\sqrt{3}$.

For the (generally noncollinear) SHG process shown in Fig. 2(b), we should satisfy the phase-matching condition $\mathbf{k}_2 = 2\mathbf{k}_1 + \mathbf{K}_{pq}$, or

$$k_2^2 = 4k_1^2 + K_{pq}^2 + 4k_1K_{pq}\cos(\alpha_{pq} - \beta_1), \quad (1)$$

where $\sin \alpha_{pq} = (p\sqrt{3}/2)(p^2 + q^2 + pq)^{-1/2}$, $k_1 = 2\pi n_1/\lambda_1$, and $k_2 = 4\pi n_2/\lambda_2$; n_1 and n_2 are the refractive indices.



Fig. 1. Schematic diagram of a 2D $\chi^{(2)}$ photonic crystal with different types of grating structures. The shaded circles mark the regions with the reverse sign of second-order susceptibility.

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Fig. 2. (a) Reciprocal lattice of the 2D crystal shown in Fig. 1; (b) SHG phase-matching wave-vector triangle.

Angle γ_1 between the fundamental frequency and second-harmonic waves [see Fig. 2(b)] is defined as $\sin \gamma_1 = (K_{qp}/k_2)\sin(\alpha_{pq} - \beta_1)$. The result of Refs. 4 and 5 can be obtained from the relation $K_{pq}^2 = (2k_1 - k_2)^2 + 8k_1k_2\sin^2(\gamma_1/2)$, which reproduces Eq. (6) of Ref. 4. For collinear SHG, we have $\alpha_{pq} = \beta_1$, and Eq. (1) leads to a simple result: $d = 4\pi\lambda(p^2 + q^2 + pq)^{1/2}/(\Delta k_2 3)$, where $\Delta k_2 = k_2 - 2k_1$.

To demonstrate the possibility of double phase matching (DPM) of two parametric processes in a 2D $\chi^{(2)}$ photonic crystal we consider an example of third-harmonic generation (THG) by simultaneous phase matching of the processes of SHG and sumfrequency mixing (SFM), $\omega + 2\omega = 3\omega$. DPM of these two processes in the SNPC of Fig. 1 is possible only when all three interacting waves are noncollinear [see Fig. 3(a)]. The first process, SHG, can be phase matched by a certain grating vector, \mathbf{K}_{pq} , and the second process, SFM, can be phase matched by another grating vector, \mathbf{K}_{mn} . Indeed, if condition (1) for SHG is fulfilled, the phase-matching condition for the SFM process is

$$K_{mn}^{2} + 2k_{12}K_{mn}\cos(\alpha_{mn} - \beta_{12}) = k_{3}^{2} - k_{12}^{2}, \quad (2)$$

with $k_{12}^2 = (k_1^2 + k_2^2 + 2k_1k_2 \cos \gamma_1)$ and $\beta_{12} = \beta_1 + \sin^{-1}[(k_2/k_{12})\sin \gamma_1]$. Solving Eqs. (1) and (2) for fixed p, q, m, n, and λ_1 , we find a unique set of parameters (β_1, d) that satisfy simultaneously the phase-matching conditions for both SHG and SFM.

Similarly, we find the DPM condition for SHG and fourth-harmonic generation (FHG), $2\omega + 2\omega = 4\omega$. The FHG process can be phase matched by grating vector \mathbf{K}_{ij} [the corresponding notation is shown in Fig. 3(b)]. Then the condition for simultaneous SHG and FHG phase matching can be found by solution of a system formed by Eq. (1) and the equation $K_{ij}^2 + 4k_2K_{ij}\cos(\alpha_{ij} - \beta_1 - \gamma_1) = k_4^2 - 4k_2^2$. DPM in a single SNPC is possible when all three in-

DPM in a single SNPC is possible when all three interacting waves are noncollinear, as that condition will reduce the corresponding harmonic efficiency. However, as will be shown below, partially collinear DPM processes are possible in an asymmetric nonlinear photonic crystal (ANPC), as shown in Fig. 4, which permits simultaneous phase matching of three different nonlinear processes. As is shown in Fig. 4, the unit cell of the ANPC is defined by three parameters: d, ϵ , and the angle δ . The magnitude and direction of the grating vectors in the ANPC are defined, respectively, as $K_{pq} = |2\pi Q_{pq}/(\epsilon d \sin \delta)|$ and $\sin \alpha_{pq} = (p/Q_{pq}) \sin \delta$, where $Q_{pq} = (p^2 + \epsilon^2 q^2 + 2\epsilon pq \cos \delta)^{1/2}$. It is important to notice that the magnitude and direction of the grating vectors in an ANPC may take any value (in contrast with the case of SNPC, where the magnitude and direction of K_{pq} can take only discrete values).

The condition for collinear SHG in an ANPC (when $\beta_1 = \alpha_{pq}$ and $\gamma_1 = 0$) is defined by the constraint that $\Delta k_2 - K_{pq} = 0$, which yields

$$d = \frac{2\pi Q_{pq}}{\Delta k_2 \epsilon \sin \delta}$$
 (3)

As above, the process of SFM, i.e., $\omega + 2\omega = 3\omega$, can be phase matched by grating vector \mathbf{K}_{mn} . Then, by noting that $K_{mn} = (Q_{mn}/Q_{pq})\Delta k_2$, we again find the phase-matching condition for the THG process:

$$\frac{k_3^2 - k_{12}^2}{(\Delta k_2)^2} - \left(\frac{Q_{mn}}{Q_{pq}}\right)^2 = \frac{2k_{12}}{\Delta k_2} \frac{Q_{mn}}{Q_{pq}} \cos \theta_{mnpq}, \quad (4)$$

where $\theta_{mnpq} \equiv (\alpha_{mn} - \alpha_{pq})$. For an arbitrary set of parameters $\{p, q, m, n\}$, Eq. (4) defines a relation between ϵ and δ that allows the DPM to be satisfied for the combined SHG–SFM process.

DPM for two processes, SHG and FHG, under the condition that both the fundamental frequency and the



Fig. 3. DPM for (a) THG, noncollinear SHG, and noncollinear SFM and (b) FHG, noncollinear SHG, and noncollinear $2\omega + 2\omega = 4\omega$ processes.



Fig. 4. Schematric diagram of an asymmetric 2D $\chi^{(2)}$ photonic crystal. For $\delta = 60^{\circ}$ and $\epsilon = 1$, it is transformed into the structure shown in Fig. 1.



Fig. 5. Examples of design parameters ϵ and d for the double- and triple-phase-matching conditions in a 2D LiNbO₃ APNC structure.

second-harmonic waves are collinear, will be achieved when both ϵ and δ satisfy the following equation:

$$\frac{k_4^2 - 4k_2^2}{(\Delta k_2)^2} - \left(\frac{Q_{ij}}{Q_{pq}}\right)^2 = \frac{4k_2}{\Delta k_2} \frac{Q_{ij}}{Q_{pq}} \cos \theta_{ijpq}.$$
 (5)

Thus an APNC permits DPM when two of the interacting waves are collinear.

The second advantage of using an APNC is the unique possibility of phase matching three parametric processes simultaneously. To illustrate this, we consider simultaneous generation of the second, third, and fourth harmonics in a single ANPC crystal. Indeed, solving the system of Eqs. (4), (5), and (3) for given p, q, m, n, i, and j gives three design parameters d, ϵ , and δ for an ANPC that support triple phase matching.

For the case when one of the indices in each pair (p, q), (i, j), and (m, n) is zero, we can obtain explicit analytical formulas. As an example, we take (p, q) =(p, 0), (m, n) = (0, n), and (i, j) = (0, j). Then, using the results obtained above, we find that $\alpha_{kl} = 0$ at k = 0 and $\alpha_{kl} = \delta$ when l = 0, and the DPM conditions for THG and FHG become

$$\cos \,\delta_{3\omega} = \frac{p^2 (k_3^2 - k_{12}^2) - \epsilon^2 n^2 (\Delta k_2)^2}{2k_{12} \Delta k_2 \epsilon n p}, \qquad (6)$$

$$\cos \delta_{4\omega} = \frac{p^2 (k_4^2 - 4k_2^2) - \epsilon^2 j^2 (\Delta k_2)^2}{4k_2 \Delta k_2 \epsilon j p} \,. \tag{7}$$

The condition for simultaneous SHG, THG, and FHG follows from $\cos \delta_{3\omega} = \cos \delta_{4\omega}$:

$$\epsilon^2 = p^2 \, rac{2j(k_3{}^2 - k_{12}{}^2)k_2 - n(k_4{}^2 - 4k_2{}^2)k_{12}}{nj(2jk_2 - nk_{12})(\Delta k_2)^2}$$

The design parameters of a 2D $\chi^{(2)}$ ANPC to satisfy double- and triple-phase-matching conditions are shown in Fig. 5 for a $LiNbO_3$ structure. We assume that all interacting waves are polarized along the zaxis, so the refractive index depends only on the wavelength. The angle $\delta_{3\omega}$ for DPM of the SHG and THG and the angle $\delta_{4\omega}$ for DPM of the SHG and FHG processes are fixed at 45°. The corresponding angle δ for simultaneous SHG, THG, and FHG depends on the fundamental wavelength and is calculated from Eq. (6) or (7).

In conclusion, we have suggested the possibility of multiple phase matching and multicolor harmonic generation in a new type of two-dimensional $\chi^{(2)}$ photonic crystal. Our results suggest that 2D photonic crystals with a periodic $\chi^{(2)}$ nonlinear response are ideal candidates for experimental observation of simultaneous generation of several harmonics and different effects associated with the multistep parametric cascading processes.

S. Saltiel's e-mail address issaltiel@phys. uni-sofia.bg.

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