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PHASE-MATCHING FOR NONLINEAR OPTICAL PARAMETRIC PROCESSES WITH MULTISTEP CASCADING

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Abstract. We present a brief overview of different methods for simultaneous phase-matching of several parametric optical processes (the so-called *multistep cascading*) in engineered structures with the modulated second-order nonlinear susceptibility. In particular, we discuss the possibility of double phase-matching in both uniform and non-uniform quasi-phase-matched (QPM) periodic optical superlattices and also in the recently fabricated two-dimensional nonlinear photonic crystals. We include also some original results demonstrating the possibility to achieve double-phase-matching with phase-reversed and periodically chirped QPM structures and also with uniform QPM structures in non-collinear geometry.

1. Introduction

Nonlinear effects produced by quadratic (or $\chi^{(2)}$) intensity-dependent response of a transparent dielectric medium are usually associated with parametric frequency conversion such as the second-harmonic generation (SHG). However, recent theoretical and experimental results demonstrate that quadratic nonlinearities can also produce many of the effects attributed to nonresonant Kerr nonlinearities via cascading of several second-order parametric processes. Such second-order cascading (SOC) effects can simulate the effective third-order processes and, in particular, those associated with the intensity-dependent change of the medium refractive index [1]. Importantly, the effective (or induced) cubic nonlinearity resulting from SOC in a quadratic medium can be of the several orders of magnitude higher than that usually measured in centrosymmetric Kerr-like nonlinear media, and it is practically instantaneous. The simplest type of SOC is based on the simultaneous action of two second-order parametric sub-processes that belong to a single second-order interaction. For example, the so-called *two-step cascading* associated with type I second-harmonic generation includes the generation of the second harmonic (SH), $\omega + \omega = 2\omega$, followed by the reconstruction of the fundamental wave through the down-conversion frequency mixing process, $2\omega - \omega = \omega$. These two sub-processes depend only on a single phase-matching parameter Δk. In particular, for the nonlinear $\chi^{(2)}$ media with a periodic modulation of the quadratic nonlinearity, i.e. the so-called *quasi-phase-matching* (QPM) structures [2], we have $\Delta k = k_2 - 2k_1 + G_m$, where $k_1 = k(\omega)$, $k_2 = k(2\omega)$ and G_m is a reciprocal vector of the periodic structure. For a homogeneous bulk $\chi^{(2)}$ medium, we have $G_m = 0$.

Multistep cascading (MSC) is another type of the SOC processes, it involves several different second-order nonlinear interactions and is characterized by at least two different phase-matching parameters [3-10]. For example, *two parent processes* of the so-called *third harmonic MSC* [3,4] are SHG, $\omega + \omega = 2\omega$, and sum-frequency mixing (SFM), $\omega + 2\omega = 3\omega$. Here, we may distinguish five harmonic *sub-processes*, namely $\omega + \omega = 2\omega$, $2\omega + \omega = 3\omega$, $3\omega - 2\omega = \omega$, $3\omega - \omega = 2\omega$, $2\omega - \omega = \omega$, and MSC results in their simultaneous action. In such a case, MSC is characterized by *two* phase-matching parameters, $\Delta k_1 = k_2 - 2k_1 + G_{m_1}$ and $\Delta k_2 = k_3 - k_2 - k_1 + G_{m_2}$, that allow to control the nonlinear interaction in a broader parameter region.

Different types of MSC processes include third harmonic MSC [3,4], two-colour MSC [5-9], fourth harmonic MSC [10], SHG and difference-frequency-generation MSC [11], etc. Various applications of MSC processes have been mentioned in the literature. In particular, MSC can support multi-component solitary waves [4,6,7,9], it usually leads to larger accumulated nonlinear phase shifts, in comparison with the simple SHG-based cascading [3,8], it can be effectively employed for the simultaneous generation of higher-order harmonics in a single quadratic crystal [12,13], and also for the generation of a cross-polarized wave [5,9] and frequency shifting in fiber optics gratings [11].

Simultaneous phase-matching of several parametric processes cannot be achieved by the traditional methods such as those based on the optical birefringence effect. However, the situation becomes different for the media with a periodic change of the sign of the quadratic nonlinearity, as occurs in the QPM structures [2] or two-dimensional (2D) $\chi^{(2)}$ nonlinear photonic crystals [14-16]. In this paper, we describe the basic principles of simultaneous phase-matching of two (or more) parametric processes in different types of 1D and 2D nonlinear optical superlattices.

2. Uniform QPM structures

In a bulk homogeneous nonlinear crystal the quadratic nonlinearity is usually constant everywhere. Several methods [17] have been suggested and employed in order to create a periodic change of the sign of the second-order nonlinear susceptibility $d^{(2)}$in QPM structures as shown in Fig. 1. From the mathematical point of view, such a periodic sequence of two domains can be described by the periodic function

$$d(z) = d_o \sum_{m \neq 0} g_m \exp(iG_m z), \qquad (1)$$

$$g_m = (2/m\pi)\sin(\pi m D), \qquad (2)$$

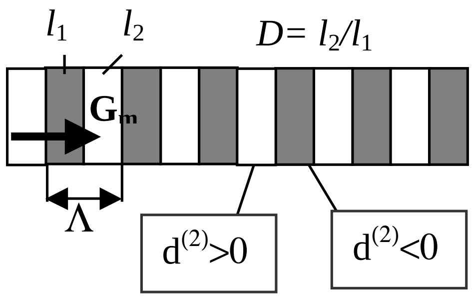

Fig. 1

where $G_m = 2\pi m/\Lambda$ is the m-th reciprocal vector of the QPM structure. The uniform QPM structure is characterized by a set of the reciprocal vectors: $\pm 2\pi/\Lambda$, $\pm 4\pi/\Lambda$, $\pm 6\pi/\Lambda$, $\pm 8\pi/\Lambda$, ..., which can be used to achieve the phase-matching conditions provided $\Delta k \rightarrow 0$. The integer number m (that can be both *positive* and *negative*) is called *the order of the wave vector phase-matching*. According to Eq. (2), the smaller is the order of the QPM reciprocal wave vector, the larger is the effective nonlinearity. If the filling factor D = 0.5, the effective quadratic nonlinearities (proportional to the parameter $d_o g_m$) that correspond to the even orders QPM vectors vanish. Importantly, such uniform QPM structures can be used for simultaneous phase-matching of two parametric processes when the interacting waves are collinear or noncollinear to the reciprocal wave vectors of the QPM structure.

A. Collinear case (two commensurable periods)

As an example, we take the *third harmonic MSC* process under the condition that the interacting waves are *collinear* to the reciprocal wave vectors of the QPM structure. We denote the mismatches of the nonlinear material without modulation of the quadratic nonlinearity ("bulk mismatches") as Δb_1 and Δb_2 (i.e., $\Delta b_1 = k_2 - 2k_1$ and $\Delta b_2 = k_3 - k_2 - k_1$) and choose the period Λ of the QPM structure in order to satisfy the phase-matching conditions $G_{m_1} = -\Delta b_1$ and $G_{m_2} = -\Delta b_2$. Two processes, SHG and SFM, are characterized by the wave-vector mismatches $\Delta k_1 = k_2 - 2k_1 + G_{m_1} = 0$ and $\Delta k_2 = k_3 - k_2 - k_1 + G_{m_2} = 0$, respectively, and they become simultaneously phase-matched for this particular choice of the QPM period. A drawback of this method is that it can satisfy simultaneously two phase-matching conditions for *discrete values* of the optical wavelength only. The values of the fundamental wavelength λ for the double phase-matching condition can be found from the relation

$$\Delta b_2/m_2 - \Delta b_1/m_1 = 0\,, \tag{3}$$

since both Δb_1 and Δb_2 are functions of the wavelength. For the third harmonic MSC process, Eq. (3) is transformed to $m_1[3n(3\lambda) - 2n(2\lambda) - n(\lambda)] - 2m_2[n(2\lambda) - n(\lambda)] = 0$, where $n(\lambda)$ is the refractive index. For a chosen pair of integer numbers (m_1, m_2) the QPM period Λ is found from the relation $\Lambda = 2\pi|m_1/\Delta b_1|$ or $\Lambda = 2\pi|m_2/\Delta b_2|$. Such a method was used for double phase-matching by several authors, (e.g. [18-21]).

B. Non-collinear case

Collinearity between the optical waves and the reciprocal vectors of the QPM structure is an important requirement for achieving a good overlapping of all the beams and a good conversion efficiency. However, phase-matching is possible even in the case when some of the waves propagate under a certain angle to the direction of the reciprocal vectors of the QPM structure, i. e. for the non-collinear case. The advantage of this method is that double phase-matching can be realized in a broad spectral range. Such a type of noncollinear interaction will be efficient for the distances corresponding to the overlap of the interacting beams.

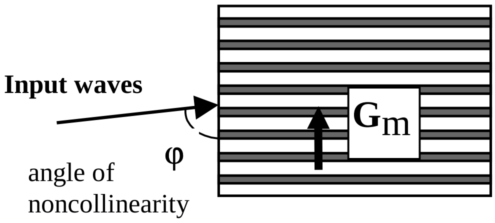

Fig. 2

Let us consider the third harmonic MSC process in this type of geometry (see Fig. 2). As has been discussed above, the simultaneous phase-matching of two parametric processes $\omega + \omega = 2\omega$ and $\omega + 2\omega = 3\omega$ is required in this case. We assume the fundamental wave at the input. As shown in Fig. 3, for the first process the phase-matching is achieved by the reciprocal vector $\mathbf{G}_{m_1}$ and the generated SH wave with wavevector $\mathbf{k}_2$ is not collinear to the fundamental wave: $2\mathbf{k}_1 + \mathbf{G}_{m_2} = \mathbf{k}_2$. The second process is phase-matched by the vector $\mathbf{G}_{m_2}$:

$\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{G}_{m_2} = \mathbf{k}_3$.

From Fig. 3 we can calculate the period of the QPM structure that allows achieving double phase-matching of the two processes,

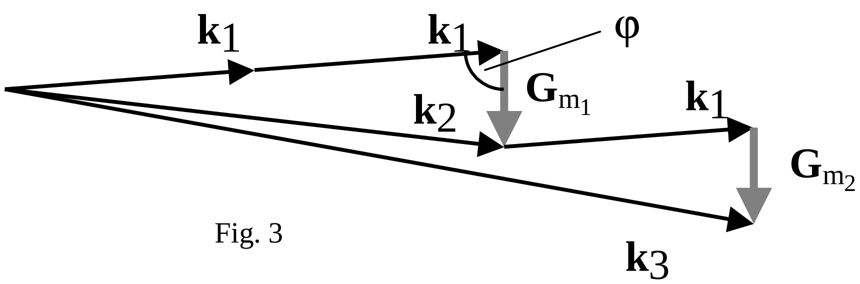

Fig. 3

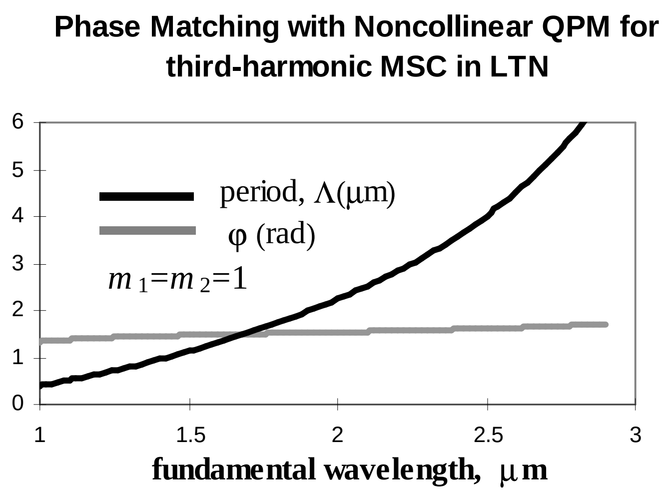

Fig. 4

$$\Lambda = 2\pi \sqrt{\frac{2(k_3^2 - 9k_1^2)m_1 - 3(k_2^2 - 4k_1^2)(m_1 + m_2)}{(m_1 + m_2)(2m_2 - m_1)m_1}} \,. \tag{4}$$

An example of double phase-matching for the third harmonic MSC process in $LiTaO_3$ for the case when all the waves are polarized along z axis of the crystal is shown in Fig. 4 $(m_1 = m_2 = 1)$.

3. Non-uniform QPM structures

Non-uniform QPM structures give also the possibility of simultaneous phase-matching of two parametric nonlinear-optical processes. We consider *three types* of such non-uniform QPM structures: *phase-reversed QPM* structures [22], periodically *chirped QPM* structures [23], and optical superlattices [12,24,25].

A. Phase-reversed QPM structures

The idea of the phase-reversed QPM structures [22] is illustrated in Fig. 5. Such a structure can be explained as a sequence of many equivalent uniform short QPM sub-structures with the length $\Lambda_{ph}/2$ connected in such a way that at the place of the joint two end layers has the same sign of the quadratic nonlinearity. Any two neighboring junctions have opposite sign of the $\chi^{(2)}$nonlinearity. By other words, the phase-reversed QPM structure is a kind of an uniform QPM structure with a change of the domain phase by π characterized by the other (larger) period Λ_{ph} .

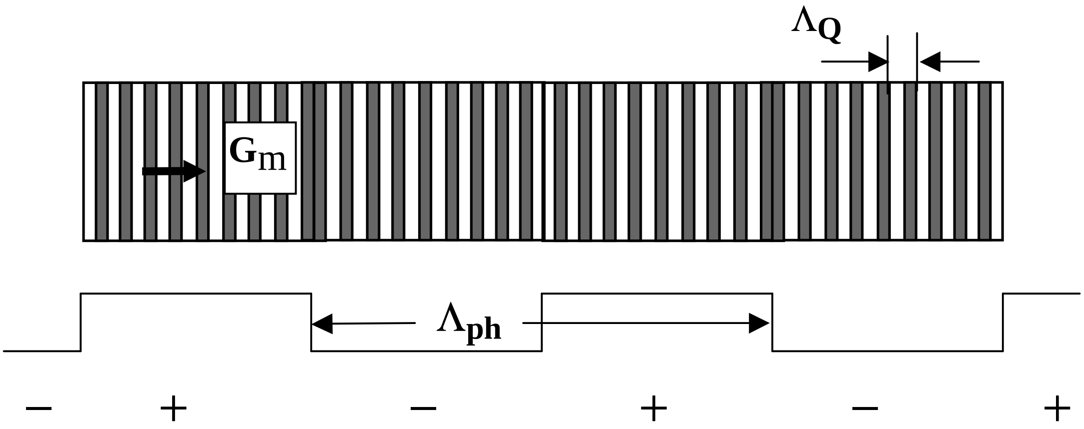

Fig. 5

The modulation of the quadratic nonlinearity $d(z)$ in the phase-reversed QPM structure with the filling factor $D = 0.5$ can be described by the response function

$$d(z) = d_o(-1)^{\mathrm{int}(2z/\Lambda_Q)}(-1)^{\mathrm{int}(2z/\Lambda_{ph})} \tag{5}$$

and can be presented in the form of the Fourier series,

$$d(z) = d_o\left[\sum_{-\infty}^{+\infty} g_l e^{(iG_l z)}\right]\left[\sum_{-\infty}^{+\infty} g_m e^{(iF_m z)}\right] = d_o\left[\sum g_{lm} e^{(iG_{lm} z)}\right], \quad l, m \neq 0 \,, \tag{6}$$

where $g_m = \dfrac{2}{m\pi}$, $g_{lm} = g_l g_m$, $G_l = \dfrac{2\pi l}{\Lambda_Q}$, $F_m = \dfrac{2\pi m}{\Lambda_{ph}}$, $G_{lm} = \dfrac{2\pi}{\Lambda_Q} l + \dfrac{2\pi}{\Lambda_{ph}} m$.

The phase-reversed QPM structure is characterized by a set of the reciprocal vectors $\{G_{lm}\}$, which are collinear to the normal of the periodic sequence with the magnitude depending on all parameters: $l, m, \Lambda_Q, \Lambda_{ph}$. Two of these vectors can be chosen to phase-match two parametric processes involved in the MSC, such that $G_{l_1 m_1} = -\Delta b_1$ and $G_{l_2 m_2} = -\Delta b_2$. Then, the two QPM periods satisfying the double-phase-matching condition are defined as:

$$\Lambda_Q = \left| \frac{2\pi(l_2 m_1 - l_1 m_2)}{m_2 \Delta b_1 - m_1 \Delta b_2} \right| \quad , \quad \Lambda_{Ph} = \left| \frac{2\pi(l_2 m_1 - l_1 m_2)}{l_2 \Delta b_1 - l_1 \Delta b_2} \right| \quad . \tag{7}$$

For the case of the third harmonic MSC Eqs. (7) are transformed into

$$\Lambda_Q = \left| \frac{\lambda(l_2 m_1 - l_1 m_2)}{(m_1 - 2m_2)n(\lambda) + 2(m_1 + m_2)n(2\lambda) - 3m_1 n(3\lambda)} \right| \quad , \tag{8}$$

$$\Lambda_{ph} = \left| \frac{\lambda(l_2 m_1 - l_1 m_2)}{(l_1 - 2l_2)n(\lambda) + 2(l_1 + l_2)n(2\lambda) - 3l_1 n(3\lambda)} \right| \quad . \tag{9}$$

In addition to Eqs (8) and (9), we should satisfy the condition that the ratio $2\Lambda_{Ph}/\Lambda_Q$ is an integer number and, therefore, this method does not allow achieving double-phase-matching for any wavelength. Nevertheless, the corresponding number of the phase-matched wavelengths is larger than that achieved in the uniform QPM structure and for collinear beams (see the method described in Sec. **2.A**).

B. Periodically chirped QPM structures

Periodically chirped QPM structures have been designed for increasing an effective (averaged) third-order nonlinearity in quadratic media [23]. The terminology “periodically chirped QPM” is used in analogy with the chirped QPM structures that has a linear increase of the period Λ_Q along the crystal.

The periodically chirped QPM structure is characterized by the QPM period Λ_Q that is a periodic function of z (see Fig. 6):

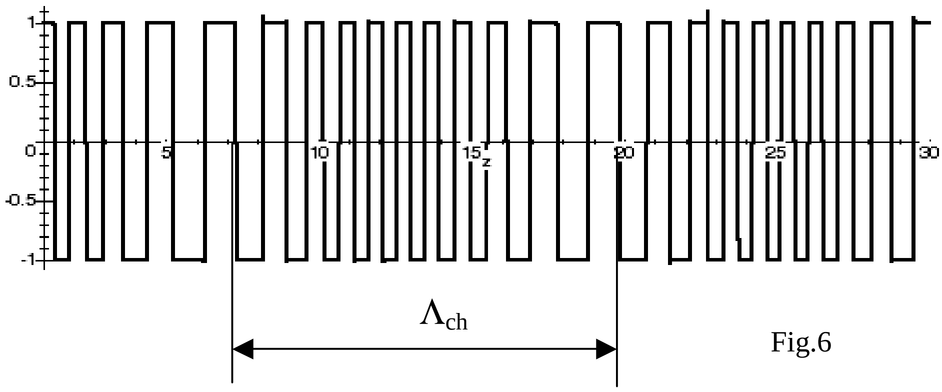

Fig.6

$$\Lambda = \Lambda_Q + \varepsilon_o \cos(2\pi z/\Lambda_{ch}). \tag{10}$$

The corresponding formulas for double phase-matching allow to find the two periods of the periodically chirped QPM structure, similar to the case of the phase-reversed QPM structure,

$$\Lambda_Q = \left|\frac{2\pi(l_2 m_1 - l_1 m_2)}{m_2 \Delta b_1 - m_1 \Delta b_2}\right| \quad , \qquad (11)$$

$$\Lambda_{ch} = \left|\frac{2\pi(l_2 m_1 - l_1 m_2)}{l_2 \Delta b_1 - l_1 \Delta b_2}\right| \quad . \qquad (12)$$

The main advantage of such periodically chirped QPM structures is the possibility to satisfy double phase-matching conditions for any wavelength in a broad spectral range (i.e. for any pair Δb_1 and Δb_2). Calculated periods for a poled $LiTaO_3$ crystal are presented in Fig.7.

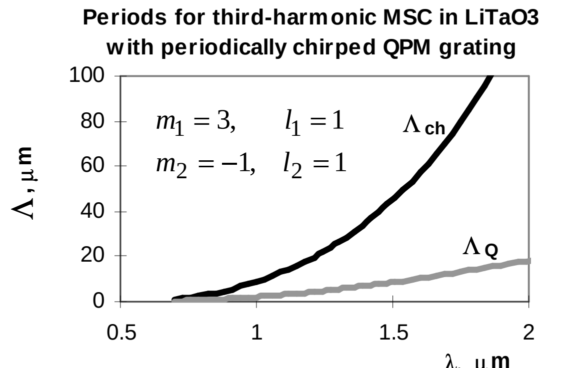

Fig. 7

C. Quasi-periodic and aperiodic optical superlattices

Another method of double-phase-matching, investigated both theoretically and experimentally is based on the use of the quasi-periodic optical superlattices (QPOS) [12,24,25] and aperiodic optical superlattices [26,27]. In the most of the cases QOPSs are built with two-component blocks (A and B) aligned in a Fibonacci (or more general quasi-periodic) sequence (see Fig. 8). Each of the blocks consists of two layers with the opposite sign of the quadratic nonlinearity. We illustrate the possibility of double-phase-matching in such a kind of structures taking, as an example, the structure consisting of two blocks aligned in a generalized sequence [25]. The modulation of the quadratic nonlinearity $d(z)$ can be described by the following Fourier expansion [28,29]:

$$d(z) = d_o \sum_{m,n} f_{m,n} \exp\left(iG_{m,n} z\right) \quad , \qquad (13)$$

where the reciprocal vectors are defined as $G_{m,n} = 2\pi\left(m + n\tau\right)/S$ and $S = \tau L_A + L_B$.

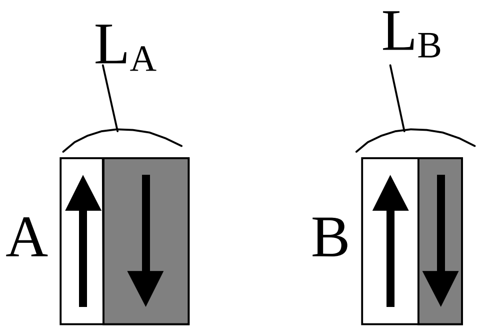

Fig. 8

For phase-matching of two processes with bulk mismatches Δb_1 and Δb_2, we solve the system of equations

$$G_{m_1,n_1} = 2\pi\left(m_1 + n_1\tau\right)/S = \Delta b_1 , \qquad (14a)$$

$$G_{m_2,n_2} = 2\pi\left(m_2 + n_2\tau\right)/S = \Delta b_2 \qquad (14b)$$

and find S and τ. Exact values for L_A, L_B and the lengths of the layers have to be found by maximizing f_{m_1,n_1} and f_{m_2,n_2}. This structure allows simultaneous phase-matching in a broad spectral range without constrains on the ratio $\Delta b_2/\Delta b_1$. Equations (14) are also valid for Fibonacci type QPOS but, since τ is fixed, double phase-matching can be satisfied for a limited number of the wavelengths [25] such that defined by $\Delta b_2/\Delta b_1 = \left(m_1 + n_1\tau\right)/\left(m_2 + n_2\tau\right)$.

In the recent papers [26,27], aperiodic optical superlattices were used for simultaneous phase-matching of several processes. In this approach, the thickness of the layers and their consequences are directly found by solving the inverse source problem maximizing the efficiency of both the processes involved into the MSC interaction.

4. 2D $\chi^{(2)}$ nonlinear photonic crystals

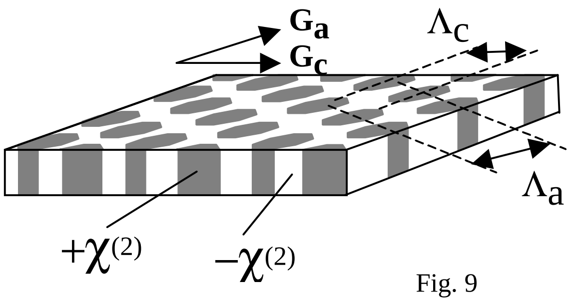

Fig. 9

Nonlinear photonic crystals (NPCs) with homogeneous (linear) refraction index and 2D periodic lattice of the quadratic nonlinear susceptibility can be effectively employed as a host media for many different types of the MSC processes [14-16]. A schematic diagram of 2D NPC is shown in Fig. 9. The simple way to obtain the phase-matching conditions for 2D NPC is to use a reciprocal lattice formed by vectors $\mathbf{G}_\mathbf{a}$ and $\mathbf{G}_\mathbf{c}$, defined as $|G_a| = 2\pi/\Lambda_a$ and $|G_c| = 2\pi/\Lambda_c$. For a hexagon lattice, we find $\Lambda_a = \Lambda_c = a\sqrt{3}/2$**,** where a is the distance between the centers of two neighboring inverted volumes. All reciprocal vectors of the 2D NPC crystal are formed by a simple rule, $\mathbf{G}_{m,n} = m\mathbf{G}_c + n\mathbf{G}_a$. Any two vectors of this set can be used to compensate for the bulk mismatches Δb_1 and Δb_2, however such phase-matching conditions can be fulfilled for noncollinear interactions only. Phase-matching conditions for the third harmonic MSC process are shown in Fig. 10: (a) for the process $\omega + \omega = 2\omega$ phase-matched by the reciprocal vector $\mathbf{G}_{m_1,n_1}$ and (b) for the process $\omega + 2\omega = 3\omega$ phase-matched by the reciprocal vector $\mathbf{G}_{m_2,n_2}$. Phase-matching is achieved by choosing the lattice spacing a and the angle of incidence β. 2D NPC can be also used for simultaneously phase-matching *of three nonlinear processes*, e.g. second, third, and fourth harmonic generation [16]. Experimentally, the simultaneous second-, third-, and fourth-harmonic generation was recently observed in poled $LiNbO_3$ 2D NPC [15].

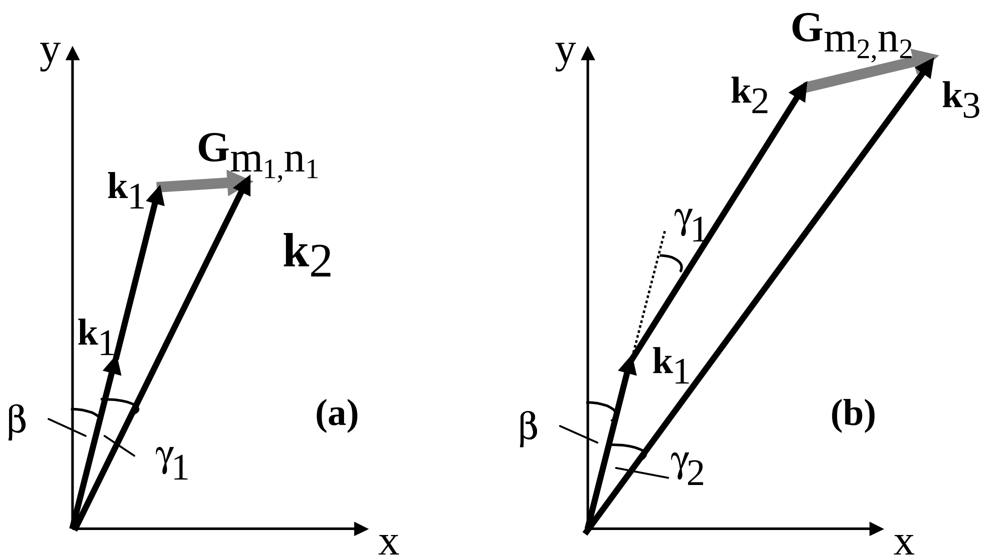

Fig. 10

5. Conclusions

We have presented a brief overview of different techniques for achieving simultaneous phase-matching of several nonlinear parametric processes in optical media with a modulated second-order nonlinear susceptibility. In all those cases, the double-phase-matched interaction is possible in a wide region of the optical wavelengths provided the QPM structure used to achieve the phase-matching conditions possesses *one extra parameter* (e.g., the modulation period in chirped QPM structures, or the second dimension, in 2D nonlinear photonic crystals). Some of the possible applications of the double-phase-matching processes include simultaneous generation of several optical frequencies in a single structure, multi-port frequency conversion, etc. The results presented above look encouraging for experimental feasibility of the predicted effects in the recently engineered 1D and 2D periodic optical superlattices.

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