Quantum Electronics **31** (10) 933 – 936 (2001)

PACS numbers: 42.65.Ky; 42.70.Qs
DOI: 10.1070/QE2001v031n10ABEH002079

Quasi-phase-matching conditions for a simultaneous generation of several harmonics of laser radiation in periodically poled crystals

S G Grechin, V G Dmitriev

***Abstract*. Quasi-phase-matching conditions are considered for a simultaneous generation of several harmonics in quadratically nonlinear periodically poled crystals.**

Keywords*: frequency conversion, periodically poled crystals, quasi-phase matching, generation of harmonics.*

Periodically poled nonlinear optical crystals provide opportunities for qualitatively new approaches towards frequency conversion of laser radiation. These include a simultaneous generation of several optical harmonics [1], second harmonic generation (SHG) simultaneously for three types of interaction [2], SHG for simultaneous realisation of phase-matching and quasi-phase-matching interactions [3], etc. In paper [4], we have mentioned the relation between various types of interactions upon SHG and the generation of third (THG) and fourth (FHG) harmonics.

In this paper, we will determine the quasi-phase-matching conditions for simultaneous SHG, THG and FHG in the case of collinear interactions in a periodically poled crystal with a quadratic nonlinearity. Because the fourth harmonic can be obtained as the sum ($\omega + 3\omega = 4\omega$) of the frequencies of the first (ω) and of the third (3ω) harmonics or by doubling ($2\omega + 2\omega = 4\omega$) the second harmonic frequency, we can write in the general case of the four processes the following expressions for the total wave detunings:

SHG: $\omega + \omega = 2\omega$,

$$\delta k_1 = k_{2i} - k_{1j} - k_{1k} + m_1 G_1 = \Delta k_1 + m_1 G_1, \qquad (1)$$

THG: $\omega + 2\omega = 3\omega$,

$$\delta k_2 = k_{3m} - k_{1k} - k_{2i} + m_2 G_2 = \Delta k_2 + m_2 G_2, \qquad (2)$$

FHG-1: $\omega + 3\omega = 4\omega$,

$$\delta k_3 = k_{4n} - k_{1j} - k_{3m} + m_3 G_3 = \Delta k_3 + m_3 G_3, \qquad (3)$$

FHG-2: $2\omega + 2\omega = 4\omega$,

$$\delta k_4 = k_{4n} - 2k_{2i} + m_4 G_4 = \Delta k_4 + m_4 G_4, \qquad (4)$$

where Δk_q are the wave detunings for a homogeneous crystal ($q = 1, 2, 3, 4$); $k_{q\alpha}$ are the wave numbers of the interacting waves; $\alpha = i, j, k, m, n$ are the indices corresponding to various types of interacting waves (o, e for uniaxial and s, f for biaxial crystals); $G_q = 2\pi\Lambda_q^{-1}$ is the wave number (pseudovector modulus) for the domain structure grating with the period Λ_q; and $m_q = 0, \pm 1, \pm 3, \pm 5, \ldots$ are the quasi-phase-matching orders.

The condition of quasi-phase matching for a particular process corresponds to $\delta k_q = 0$ (in this case, the 'traditional' phase-matching condition $\Delta k_q = 0$ corresponds to $m_q = 0$), and a simultaneous quasi-phase matching for all the four processes may occur in the same domain structure (i.e., for $G_1 = G_2 = G_3 = G_4$) in the general case for different orders of quasi-phase matching or, in other words, for different coherence lengths $L_{\rm coh}^{(q)} = \pi/\Delta k_q = \Lambda_q/(2m_q)$.

Let us assume that the quasi-phase-matching conditions $\delta k_1 = \delta k_2 = 0$ are satisfied for SHG and THG in the same domain structure ($G_1 = G_2$) and for the same order of quasi-phase matching ($m_1 = m_2$). Then, we have

$$2k_{2i} = k_{1j} + k_{3m}. \qquad (5)$$

It follows from Eqns (3) – (5) that in this case $\Delta k_3 = \Delta k_4$. If the quasi-phase-matching condition is satisfied, for example, for FHG-1 ($\delta k_3 = 0$), we obtain $\delta k_4 = m_4 G_4 - m_3 G_3$. We assume that the quasi-phase-matching condition for FHG-2 ($\delta k_4 = 0$) will be provided by the equality $m_4 G_4 = m_3 G_3$ for the same phase-matching order ($m_3 = m_4$), and, hence, for the same domain structure ($G_3 = G_4$). In the general case, we have $m_1 \neq m_3$ and $G_1 \neq G_3$.

This is illustrated in Fig. 1 showing the dependence of the coherence length $L_{\rm coh}^{(q)}$ on the wavelength of the fundamental (laser) radiation for all four collinear eee processes for radiation propagating in the xy plane of a stoichiometric $LiNbO_3$ crystal, for which the coefficients of the Sellmeyer equation are borrowed from Ref. [5]. Fig. 1 shows that pairwise equality of the coherence lengths is observed at the wavelengths $\lambda_1 = 3579.54$ nm and $\lambda_2 = 4256.45$ nm: for the wavelength λ_1, we have $L_{\rm coh}^{(1)} = L_{\rm coh}^{(2)} = 16.05$ μm and $L_{\rm coh}^{(3)} = L_{\rm coh}^{(4)} = 13.62$ μm, while for the wavelength λ_2, we have $L_{\rm coh}^{(1)} = L_{\rm coh}^{(3)} = 14.09$ μm and $L_{\rm coh}^{(2)} = L_{\rm coh}^{(4)} = 15.85$ μm.

For a simultaneous fulfilment of the quasi-phase-matching conditions at λ_1 for SHG and THG, it is sufficient to choose the domain structure period $\Lambda_1 = 2m_1 L_{\rm coh}^{(1)}$, while the

S G Grechin Scientific-Research Institute of Radioelectronics and Laser Technology at the N E Bauman Moscow State Technical University, 2ya Baumanskaya ul. 5, 107005 Moscow, Russia; e-mail: gera@mx.bmstu.ru;
V G Dmitriev M F Stel'makh Scientific-Research Institute 'Polyus' (State Enterprise), ul. Vvedenskogo 3, 117342 Moscow, Russia; e-mail: vgdmitr@orc.ru

Received 14 June 2001
Kvantovaya Elektronika **31** (10) 933 – 936 (2001)
Translated by Ram Wadhwa

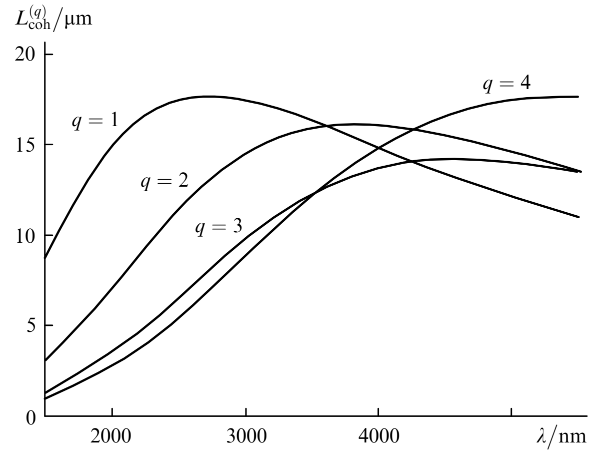

Figure 1. Dispersion curves for the coherence lengths of collinear eee processes of SHG ($q = 1$), THG ($q = 2$), FHG-1 ($q = 3$) and FHG-2 ($q = 4$) in a stoichiometric $LiNbO_3$ crystal. Pairwise equality of coherence lengths is observed at wavelengths $\lambda_1 = 3579.54$ nm and $\lambda_2 = 4256.45$ nm.

phase-matching condition for FHG-1 and FHG-2 is realised simultaneously for the domain structure period $\Lambda_3 = 2m_3 L_{\rm coh}^{(3)}$. For a simultaneous fulfilment of the quasi-phase-matching conditions for all four processes in the same domain structure, the equality $\Lambda_1 = \Lambda_3$ must be provided:

$$\Lambda_1 = \frac{2\pi}{G_1} = 2m_1 L_{\rm coh}^{(1)} = 2m_3 L_{\rm coh}^{(3)}, \tag{6}$$

i.e., the domain structure period must be a multiple of two coherence lengths simultaneously. It follows from (6) that

$$\frac{L_{\rm coh}^{(1)}}{L_{\rm coh}^{(3)}} = \frac{m_3}{m_1}, \tag{7}$$

i.e., the ratio of the coherence lengths must be equal to an odd number or a ratio of odd integers. Expression (7) can be used to determine the conditions (i.e., the wavelengths of the fundamental radiation and the types of interacting waves) under which the quasi-phase-matching conditions are satisfied for all four processes (1)–(4), which restricts the number of combinations of the types of interaction (there are 64 such combinations, but, first, the quasi-phase-matching condition is obviously not satisfied for all pairs of interactions in the transparency region of the crystal and, second, condition (7) is also not satisfied for all of them).

In analogy with the condition for λ_1, we obtain the following relation when the quasi-phase-matching conditions for λ_2 are satisfied simultaneously for SHG and FHG-1: $\Lambda_1 = 2m_1 L_{\rm coh}^{(1)}$. The corresponding relation for THG and FHG-2 is $\Lambda_2 = 2m_2 L_{\rm coh}^{(2)}$. For a simultaneous fulfilment of the quasi-phase-matching conditions for all four processes in the same domain structure, the equality $\Lambda_1 = \Lambda_2$ should be satisfied, i.e.,

$$\Lambda_1 = \frac{2\pi}{G_1} = 2m_1 L_{\rm coh}^{(1)} = 2m_2 L_{\rm coh}^{(2)}, \tag{8}$$

whence

$$\frac{L_{\rm coh}^{(1)}}{L_{\rm coh}^{(2)}} = \frac{m_2}{m_1}. \tag{9}$$

Let us illustrate the above by the example of a negative $LiNbO_3$ uniaxial crystal of a stoichiometric composition for radiation propagating in the xy plane. Table 1 presents the results of computation of the coherence length for various combinations of the types of interaction for all four processes (1)–(4). The first four columns of Table 1 show the types of interaction considered by us, while the fifth column contains the wavelengths at which the coherence lengths coincide pairwise for processes (1) and (2), i.e., $L_{\rm coh}^{(1,2)}$, and for processes (3) and (4), i.e., $L_{\rm coh}^{(3,4)}$. The values of $L_{\rm coh}^{(3,4)}$ and $L_{\rm coh}^{(1,2)}$ are presented in the sixth and seventh columns, while the eighth column contains the values of the ratio $L_{\rm coh}^{(1,2)}/L_{\rm coh}^{(3,4)}$: if this ratio is equal to an odd integer or to the ratio of odd integers, quasi-phase matching is realised simultaneously for all four processes: SHG, THG, FHG-1 and FHG-2.

The phase-matching order m in Eqns (1)–(4) is an odd integer and may be positive or negative (the latter corresponds to the negative coherence length or, in other words, to a direction opposite to the direction of the pseudovector $\boldsymbol{G}_q$ of the domain structure). The mutual relation between the two pairs of processes (SHG, THG) and (FHG-1, FHG-2) in Eqs (1)–(4) shows that for the quasi-phase-mat-

Table 1.

$2\omega + 2\omega = 4\omega$	$\omega + 3\omega = 4\omega$	$\omega + 2\omega = 3\omega$	$\omega + \omega = 2\omega$	λ/nm	$L_{\rm coh}^{(3,4)}/\mu$m	$L_{\rm coh}^{(1,2)}/\mu$m	$L_{\rm coh}^{(1,2)}/L_{\rm coh}^{(3,4)}$
eeo	eeo	eee	eee	3579.54	3.95	16.05	4.07
		oee	oee	3579.54	3.95	35.51	9.00
	oeo	eee	oee	5349.02	5.97	13.94	2.34
		oee	ooe	5349.02	5.97	18.01	3.02
ooo	eoo	eoo	eeo	2144.32	3.00	4.89	1.63
		ooo	oeo	2144.32	3.00	7.19	3.40
	ooo	eoo	oeo	3541.2	10.61	9.21	0.87
		ooo	ooo	3541.2	10.61	13.53	1.27
eee	eee	oee	oee	3579.54	12.62	35.51	2.81
		eee	eee	3579.54	12.62	16.05	1.27
	oee	oee	ooe	5349.05	17.69	18.01	1.02
		eee	oee	5349.05	17.69	13.94	0.79
ooe	eoe	eoo	eeo	2144.32	339.45	4.89	0.01
		ooo	eeo	2144.32	339.45	7.19	0.02
	ooe	eoo	oeo	3541.2	−12.19	9.21	−0.76
		ooo	ooo	3541.2	−12.19	13.53	−1.11

ching condition to be realised simultaneously for all processes, the signs of the quasi-phase-matching orders for the pairs of processes (1), (2) and (3), (4) must be the same within these pairs, although the pairs may have opposite signs. Note that in the last two rows of Table 1, the pairs of processes (1), (2) and (3), (4) have different signs of the quasi-phase-matching orders.

If we do not set out to fulfil the quasi-phase-matching conditions simultaneously for all four processes, the quasi-phase-matching orders may have opposite signs in the pairs of processes (SHG, THG) and (FHG-1, FHG-2). Table 2 shows the results of computations of the coherence length for the same stoichiometric $LiNbO_3$ crystal in which the processes FHG-1 and FHG-2 are realised simultaneously when the first (ω), second (2ω) and third (3ω) harmonics are supplied at the input of a nonlinear periodically poled crystal. Naturally, a simultaneous realisation of quasi-phase-matching conditions for SHG and THG cannot be attained for any of the processes shown in Table 2.

Table 2.

$2\omega + 2\omega = 4\omega$	$\omega + 3\omega = 4\omega$	λ/nm	$L_{\mathrm{coh}}^{(3)}$/μm	$L_{\mathrm{coh}}^{(4)}$/μm
eee	ooe	3439.5	11.76	−11.76
		4003.4	14.84	−14.84
ooe	ooe	2027.67	24.66	−24.66
	oee	2725.8	−13.49	13.49
		5002.18	−19.21	19.21
	eoe	2157.38	−1873.04	1873.04
	eee	3424.66	−12.01	12.01
		4132.4	−13.95	13.95

Similar computations can be made for the first pair of processes (SHG, THG) for the same and opposite signs of the coherence lengths. The realisation of quasi-phase matching (SHG and THG simultaneously) for FHG-1 and FHG-2 is ruled out. This method can be used to eliminate undesirable competition from the other pair of processes.

Note that if the quasi-phase-matching orders (coherence lengths) have opposite signs, the signs of the effective nonlinearity d_{eff} of the crystal in the pairs will also be different for these pairs. In the case of pairs of processes FHG-1 and FHG-2, this leads to a decrease in the total efficiency of conversion to the fourth harmonic. For this reason, it is important that the signs of d_{eff} be identical for both processes (e.g., as a result of a proper selection of the octant of interaction). This will increase the FHG conversion efficiency, as was shown for SHG in our paper [6].

Consider now the possibility of realising quasi-phase matching simultaneously in all four processes at the same domain structure for different quasi-phase-matching orders in each pair of processes. Let us assume, for example, that the conditions of simultaneous phase-matching are satisfied for $m_1 \neq m_2$ and $G_1 = G_2 = G$ for processes (1) and (2), i.e., SHG and THG. In this case, if the condition $\delta k_3 = 0$ is satisfied simultaneously in the same structure (for FHG-1), we obtain from Eqns (1)–(4) the relation $\delta k_4 = -(m_2 - m_1)G + m_4 G_4 - m_3 G)$ (because $G_3 = G$ in this case), and quasi-phase matching is possible for FHG-2 ($\delta k_4 = 0$) in the same structure ($G_4 = G$) under the condition

$$m_1 + m_4 = m_2 + m_3. \quad (10)$$

This relation describes the possibility of realising quasi-phase-matching simultaneously for all four processes (1)–(4) in the same domain structure.

The stated above is illustrated in Fig. 2 which shows the dependences of the required domain structure period $\Lambda_q = 2L_{\mathrm{d}}^{(q)}$, where L_{d} is the domain width, on the wavelength of the fundamental radiation in a biaxial KTP crystal for four processes (1)–(4) with different quasi-phase-matching orders selected according to relation (10) ($m_1 = 3$, $m_2 = 5$, $m_3 = 9$, $m_4 = 11$, so that $m_1 + m_4 = m_2 + m_3 = 14$) for the sss interaction in the xy plane (recall that the indices o and e should be replaced by s and f on passing from uniaxial to biaxial crystals).

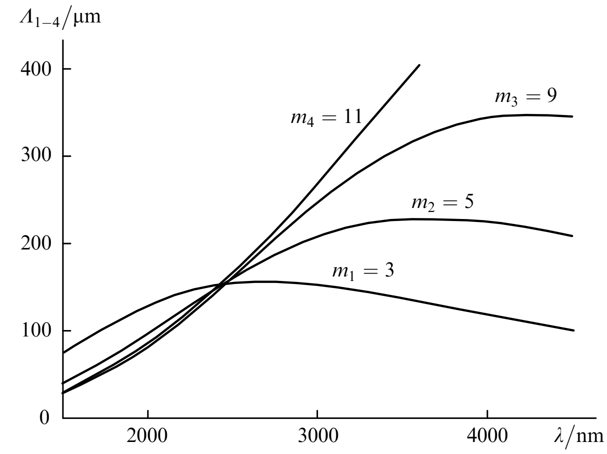

Figure 2. Dispersion curves for the domain structure period required for the occurrence of collinear sss processes of SHG ($m_1 = 3$), THG ($m_2 = 5$), FHG-1 ($m_3 = 9$) and FHG-2 ($m_4 = 11$) in a KTP crystal. Intersection of the curves for $\Lambda_{1-4} \simeq 153$ μm ($L_{\mathrm{d}} = 76.46$ μm) is observed at the wavelength 2445.5 nm.

Calculations were made by using the coefficients of Sellmeyer equation from [7]. The conditions of simultaneous quasi-phase matching for processes (1)–(4) at the fundamental radiation wavelength 2442.5 nm for $\Lambda_{1-4} \simeq 153$ μm were realised for various quasi-phase-matching orders and crystal heating to 31.5 °C.

The obtained results can also be generalised to the generation of higher-order harmonics. Thus, if the quasi-phase-matching condition $\delta k_q = 0$ is satisfied in Eqns (1)–(4), premises are automatically created for a simultaneous generation of the fifth harmonic (FiHG) in processes $\omega + 4\omega = 5\omega$ (FiHG-1) and $2\omega + 3\omega = 5\omega$ (FiHG-2):

$$\text{FiHG-1:} \quad \delta k_5 = k_{5s} - k_{1k} - k_{4n} + m_5 G_5 = \Delta k_5 + m_5 G_5, \quad (11)$$

$$\text{FiHG-2:} \quad \delta k_6 = k_{5s} - k_{2i} - k_{3m} + m_6 G_6 = \Delta k_6 + m_6 G_6. \quad (12)$$

For $\Delta k_1 = \Delta k_4$, we obtain $\Delta k_5 = \Delta k_6$.

Similarly, for the sixth harmonic generation (SiHG) in the processes $\omega + 5\omega = 6\omega$ (SiHG-1), $2\omega + 4\omega = 6\omega$ (SiHG-2) and $3\omega + 3\omega = 6\omega$ (SiHG-3), we have

$$\text{(SiHG-1):} \ \delta k_7 = k_{6p} - k_{5s} - k_{1j} + m_7 G_7 = \Delta k_7 + m_7 G_7, \quad (13)$$

$$\text{(SiHG-2):} \ \delta k_8 = k_{6p} - k_{2i} - k_{4n} + m_8 G_8 = \Delta k_8 + m_8 G_8, \quad (14)$$

$$\text{(SiHG-3):} \ \delta k_9 = k_{6p} - 2k_{3m} + m_9 G_9 = \Delta k_9 + m_9 G_9. \quad (15)$$

It follows from Eqns (13) and (15) that if the quasi-phase-matching condition is satisfied for all interactions at the same order, the following equality holds:

$$k_{4n} + k_{2i} = k_{5s} + k_{1j} \,.$$

This equality creates premises for the fulfilment of quasi-phase-matching conditions for SHG (1) and FiHG-1 (11).

In this work, we have confined our analysis to the fulfilment of quasi-phase-matching conditions without paying attention to the effective nonlinearity. In particular, for crystals of point group $3m$ in the xy plane, effective nonlinearities for the following types of interaction are nonzero: eee, ooe, ooo, and eoo. Upon a change in the directions of the wave vectors of interacting waves $\boldsymbol{k}_q(\omega_i)$ (angle θ), the effective nonlinearities are nonzero and all the processes can be realised.

Thus, there exists a connection between the quasi-phase-matching orders for the simultaneous generation of various harmonics of laser radiation in periodically poled crystals. This allows the creation of multifrequency converters of laser radiation into harmonics, which can find a number of interesting applications, e.g., for fabricating a source of electromagnetic shock waves in the optical range. However, if, for example, only second and third harmonics are required, a simultaneous generation of FHG may become a competing process, preventing the required conversion and reducing its efficiency. The undesirable pair of processes can be suppressed by using interactions with opposite signs of the quasi-phase-matching orders.

All the results presented in this work were obtained by using the reference-computational packet of applied programs LID-SHG (Laser Investigator and Designer – Second Harmonic Generation) located at the site http://www.bmstu.ru/~lid.

Acknowledgements. This work was supported by the Russian Foundation for Basic Research (Grant No. 00-02-17857).

References

1. Fejer M M, Magel G A, Jundt D H, Byer R L *IEEE J. Quantum Electron.* **28** 2631 (1992)
2. Grechin S G, Dmitriev V G *Kvantovaya Elektron.* **26** 151 (1999) [*Quantum Electron.* **29** 151 (1999)]
3. Grechin S G, Dmitriev V G, Yur'ev Yu V *Kvantovaya Elektron.* **26** 155 (1999) [*Quantum Electron.* **29** 155 (1999)]
4. Dmitriev V G, Grechin S G *Proc. SPIE-Int. Soc. Opt. Eng.* **3733** 228 (2001)
5. Smith D S, Riccius H D, Edwin R P *Opt. Commun.* **17** 332 (1976)
6. Grechin S G, Dmitriev V G *Kvantovaya Elektron.* **31** 929 (2001) [*Quantum Electron.* **31** 929 (2001)]
7. Anthon D W, Crowder C D *Appl. Opt.* **27** 2650 (1988)