

## 1.7. NONLINEAR OPTICAL PROPERTIES

If ABDP relations, defined in Section 1.7.2.2.1, are verified, then the three tensorial contractions in equations (1.7.3.22) are equal to the same quantity, which we write  $\varepsilon_0\chi_{\text{eff}}^{(2)}$ , where  $\chi_{\text{eff}}^{(2)}$  is called the effective coefficient:

$$\begin{aligned}\chi_{\text{eff}}^{(2)} &= \mathbf{e}_1 \cdot \chi^{(2)}(\omega_1 = \omega_3 - \omega_2) \cdot \mathbf{e}_3 \otimes \mathbf{e}_2 \\ &= \mathbf{e}_2 \cdot \chi^{(2)}(\omega_2 = \omega_3 - \omega_1) \cdot \mathbf{e}_3 \otimes \mathbf{e}_1 \\ &= \mathbf{e}_3 \cdot \chi^{(2)}(\omega_3 = \omega_1 + \omega_2) \cdot \mathbf{e}_1 \otimes \mathbf{e}_2.\end{aligned}\quad (1.7.3.23)$$

The same considerations lead to the same kind of equations for a four-wave interaction:

$$\begin{aligned}\frac{\partial E_1(X_1, Y_1, Z)}{\partial Z} &= j\kappa_1\varepsilon_0\chi_{\text{eff}}^{(3)}E_4(X_4, Y_4, Z)E_2^*(X_2, Y_2, Z) \\ &\quad \times E_3^*(X_3, Y_3, Z)\exp(j\Delta kZ) \\ \frac{\partial E_2(X_2, Y_2, Z)}{\partial Z} &= j\kappa_2\varepsilon_0\chi_{\text{eff}}^{(3)}E_4(X_4, Y_4, Z)E_1^*(X_1, Y_1, Z) \\ &\quad \times E_3^*(X_3, Y_3, Z)\exp(j\Delta kZ) \\ \frac{\partial E_3(X_3, Y_3, Z)}{\partial Z} &= j\kappa_3\varepsilon_0\chi_{\text{eff}}^{(3)}E_4(X_4, Y_4, Z)E_1^*(X_1, Y_1, Z) \\ &\quad \times E_2^*(X_2, Y_2, Z)\exp(j\Delta kZ) \\ \frac{\partial E_4(X_4, Y_4, Z)}{\partial Z} &= j\kappa_4\varepsilon_0\chi_{\text{eff}}^{(3)}E_1(X_1, Y_1, Z)E_2(X_2, Y_2, Z) \\ &\quad \times E_3(X_3, Y_3, Z)\exp(-j\Delta kZ).\end{aligned}\quad (1.7.3.24)$$

The conventions of notation are the same as previously and the phase mismatch is  $\Delta k = k(\omega_4) - [k(\omega_1) + k(\omega_2) + k(\omega_3)]$ . The effective coefficient is

$$\begin{aligned}\chi_{\text{eff}}^{(3)} &= \mathbf{e}_1 \cdot \chi^{(3)}(\omega_1 = \omega_4 - \omega_2 - \omega_3) \cdot \mathbf{e}_4 \otimes \mathbf{e}_2 \otimes \mathbf{e}_3 \\ &= \mathbf{e}_2 \cdot \chi^{(3)}(\omega_2 = \omega_4 - \omega_1 - \omega_3) \cdot \mathbf{e}_4 \otimes \mathbf{e}_1 \otimes \mathbf{e}_3 \\ &= \mathbf{e}_3 \cdot \chi^{(3)}(\omega_3 = \omega_4 - \omega_1 - \omega_2) \cdot \mathbf{e}_4 \otimes \mathbf{e}_1 \otimes \mathbf{e}_2 \\ &= \mathbf{e}_4 \cdot \chi^{(3)}(\omega_4 = \omega_1 + \omega_2 + \omega_3) \cdot \mathbf{e}_1 \otimes \mathbf{e}_2 \otimes \mathbf{e}_3.\end{aligned}\quad (1.7.3.25)$$

Expressions (1.7.3.23) for  $\chi_{\text{eff}}^{(2)}$  and (1.7.3.25) for  $\chi_{\text{eff}}^{(3)}$  can be condensed by introducing adequate third- and fourth-rank tensors to be contracted, respectively, with  $\chi^{(2)}$  and  $\chi^{(3)}$ . For example,  $\chi_{\text{eff}}^{(2)} = \chi^{(2)} \cdot \mathbf{e}_3 \otimes \mathbf{e}_1 \otimes \mathbf{e}_2$  or  $\chi_{\text{eff}}^{(3)} = \chi^{(3)} \cdot \mathbf{e}_4 \otimes \mathbf{e}_1 \otimes \mathbf{e}_2 \otimes \mathbf{e}_3$ , and similar expressions. By substituting (1.7.3.8) in (1.7.3.22), we obtain the derivatives of Manley–Rowe relations (1.7.2.40)  $\partial N(\omega_3, Z)/\partial Z = -\partial N(\omega_k, Z)/\partial Z$  ( $k = 1, 2$ ) for a three-wave mixing, where  $N(\omega_i, Z)$  is the  $Z$  photon flow. Identically with (1.7.3.24), we have  $\partial N(\omega_4, Z)/\partial Z = -\partial N(\omega_k, Z)/\partial Z$  ( $k = 1, 2, 3$ ) for a four-wave mixing.

In the general case, the nonlinear polarization wave and the generated wave travel at different phase velocities,  $(\omega_1 + \omega_2)/[k(\omega_1) + k(\omega_2)]$  and  $\omega_3/[k(\omega_3)]$ , respectively, because of the frequency dispersion of the refractive indices in the crystal. Then the work per unit time  $W(\omega_i)$ , given in (1.7.2.39), which is done on the generated wave  $\mathbf{E}(\omega_i, Z)$  by the nonlinear polar-

ization  $\mathbf{P}^{\text{NL}}(\omega_i, Z)$ , alternates in sign for each phase shift of  $\pi$  during the  $Z$ -propagation, which leads to a reversal of the energy flow (Bloembergen, 1965). The length leading to the phase shift of  $\pi$  is called the coherence length,  $L_c = \pi/\Delta k$ , where  $\Delta k$  is the phase mismatch given by (1.7.3.22) or (1.7.3.24).

## 1.7.3.2.2. Phase matching

The transfer of energy between the waves is maximum for  $\Delta k = 0$ , which defines phase matching: the energy flow does not alternate in sign and the generated field grows continuously. Note that a condition relative to the phases  $\Phi(\omega_i, Z)$  also exists: the work of  $\mathbf{P}^{\text{NL}}(\omega_i, Z)$  on  $\mathbf{E}(\omega_i, Z)$  is maximum if these two waves are  $\pi/2$  out of phase, that is to say if  $\Delta kZ + \Delta\Phi(Z) = \pi/2$ , where  $\Delta\Phi(Z) = \Phi(\omega_3, Z) - [\Phi(\omega_1, Z) + \Phi(\omega_2, Z)]$ ; thus in the case of phase matching, the phase relation is  $\Phi(\omega_3, Z) = \Phi(\omega_1, Z) + \Phi(\omega_2, Z) + \pi/2$  (Armstrong *et al.*, 1962). The complete initial phase matching is necessarily achieved when at least one wave among all the interacting waves is not incident but is generated inside the nonlinear crystal: in this case, its initial phase is locked on the good one. Phase matching is usually realized by the matching of the refractive indices using birefringence of anisotropic media as it is studied here. From the point of view of the quantum theory of light, the phase matching of the waves corresponds to the total photon-momentum conservation *i.e.*

$$\sum_{i=1}^{\gamma-1} \hbar k(\omega_i) = \hbar k(\omega_\gamma) \quad (1.7.3.26)$$

with  $\gamma = 3$  for a three-photon interaction and  $\gamma = 4$  for a four-photon interaction.

According to (1.7.3.4), the phase-matching condition (1.7.3.26) is expressed as a function of the refractive indices in the direction of propagation considered ( $\theta, \varphi$ ); for an interaction where the  $\gamma$  wavevectors are collinear, it is written

$$\sum_{i=1}^{\gamma-1} \omega_i n(\omega_i, \theta, \varphi) = \omega_\gamma n(\omega_\gamma, \theta, \varphi) \quad (1.7.3.27)$$

with

$$\sum_{i=1}^{\gamma-1} \omega_i = \omega_\gamma. \quad (1.7.3.28)$$

(1.7.3.28) is the relation of the energy conservation.

The efficiency of a nonlinear crystal directly depends on the existence of phase-matching directions. We shall see by considering in detail the effective coefficient that phase matching is a necessary but insufficient condition for the best expression of the nonlinear optical properties.

In an hypothetical non-dispersive medium [ $\partial n(\omega)/\partial\omega = 0$ ], (1.7.3.27) is always verified for each of the eigen refractive indices  $n^+$  or  $n^-$ ; then any direction of propagation is a phase-matching direction. In a dispersive medium, phase matching can be achieved only if the direction of propagation has a birefringence which compensates the dispersion. Except for a propagation along the optic axis, there are two possible values,  $n^+$  and  $n^-$  given by (1.7.3.6), for each of the three or four refractive indices involved in the phase-matching relations, that is to say  $2^3$  or  $2^4$  possible combinations of refractive indices for a three-wave or a four-wave process, respectively.

For a three-wave process, only three combinations among the  $2^3$  are compatible with the dispersion in frequency (1.7.3.7) and with the momentum and energy conservations (1.7.3.27) and (1.7.3.28). Thus the phase matching of a

Table 1.7.3.1. Correspondence between the phase-matching relations, the configurations of polarization and the types according to the sum- and difference-frequency generation processes SFG ( $\omega_3 = \omega_1 + \omega_2$ ), DFG ( $\omega_1 = \omega_3 - \omega_2$ ) and DFG ( $\omega_2 = \omega_3 - \omega_1$ )

$\mathbf{e}^\pm$  are the unit electric field vectors relative to the refractive indices  $n^\pm$  in the phase-matching direction (Boulanger & Marnier, 1991).

Phase-matching relations	Configurations of polarization			Types of interaction		
	$\omega_3$	$\omega_1$	$\omega_2$	SFG ( $\omega_3$ )	DFG ( $\omega_1$ )	DFG ( $\omega_2$ )
$\omega_3 n_3^- = \omega_1 n_1^+ = \omega_2 n_2^+$	$\mathbf{e}^-$	$\mathbf{e}^+$	$\mathbf{e}^+$	I	II	III
$\omega_3 n_3^- = \omega_1 n_1^- = \omega_2 n_2^+$	$\mathbf{e}^-$	$\mathbf{e}^-$	$\mathbf{e}^+$	II	III	I
$\omega_3 n_3^- = \omega_1 n_1^+ = \omega_2 n_2^-$	$\mathbf{e}^-$	$\mathbf{e}^+$	$\mathbf{e}^-$	III	I	II

# 1. TENSORIAL ASPECTS OF PHYSICAL PROPERTIES

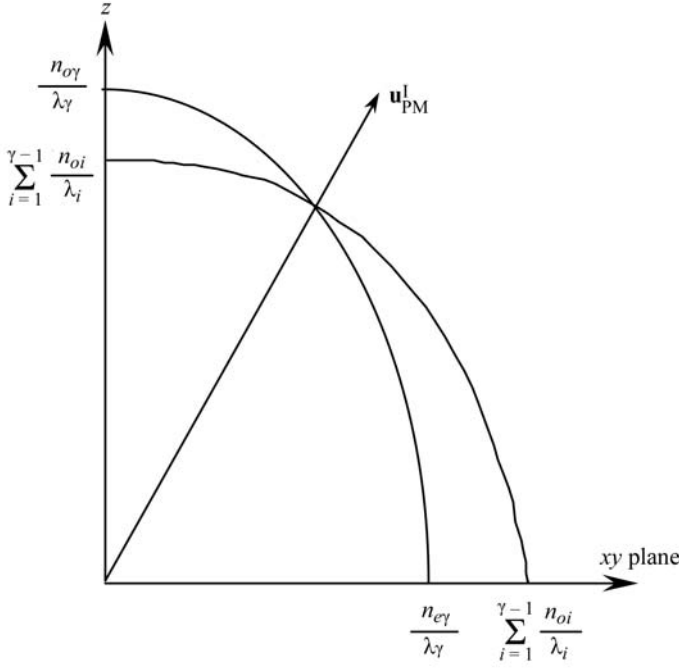


Fig. 1.7.3.4. Index surface sections in a plane containing the optic axis  $z$  of a negative uniaxial crystal allowing collinear type-I phase matching for SFG ( $\omega_3 = \omega_1 + \omega_2$ ),  $\gamma = 3$ , or for SFG ( $\omega_4 = \omega_1 + \omega_2 + \omega_3$ ),  $\gamma = 4$ .  $\mathbf{u}_{\text{PM}}^{\text{I}}$  is the corresponding phase-matching direction.

three-wave interaction is allowed for three configurations of polarization given in Table 1.7.3.1.

The designation of the type of phase matching, I, II or III, is defined according to the polarization states at the frequencies which are added or subtracted. Type I characterizes interactions for which these two waves are identically polarized; the two corresponding polarizations are different for types II and III. Note that each phase-matching relation corresponds to one sum-frequency generation SFG ( $\omega_3 = \omega_1 + \omega_2$ ) and two difference-frequency generation processes, DFG ( $\omega_1 = \omega_3 - \omega_2$ ) and DFG ( $\omega_2 = \omega_3 - \omega_1$ ). Types II and III are equivalent for SHG because  $\omega_1 = \omega_2$ .

For a four-wave process, only seven combinations of refractive indices allow phase matching in the case of normal dispersion; they are given in Table 1.7.3.2 with the corresponding configurations of polarization and types of SFG and DFG.

The convention of designation of the types is the same as for three-wave interactions for the situations where one polarization state is different from the three others, leading to the types I, II, III and IV. The criterion corresponding to type I cannot be applied to the three other phase-matching relations where two waves have the same polarization state, different from the two others. In this case, it is convenient to refer to each phase-matching relation by the same roman numeral, but with a different index:  $\text{V}^i$ ,  $\text{VI}^i$  and  $\text{VII}^i$ , with the index  $i = 1, 2, 3, 4$  corresponding to the index of the frequency generated by the SFG or DFG. For THG ( $\omega_1 = \omega_2 = \omega_3$ ), types II, III and IV are equivalent, and so are types  $\text{V}^4$ ,  $\text{VI}^4$  and  $\text{VII}^4$ .

The index surface allows the geometrical determination of the phase-matching directions, which depend on the relative ellipticity of the internal (–) and external (+) sheets divided by the corresponding wavelengths: according to Tables 1.7.3.1 and 1.7.3.2 the directions are given by the intersection of the internal sheet of the lowest wavelength  $[n^-(\lambda_\gamma, \theta, \varphi)]/(\lambda_\gamma)$  with a linear combination of the internal and external sheets at the other frequencies  $\sum_{i=1}^{\gamma-1} [n^\pm(\lambda_i, \theta, \varphi)]/(\lambda_i)$ . The existence and loci of these intersections depend on specific inequalities between the principal refractive indices at the different wavelengths. Note that independently of phase-matching considerations, normal dispersion and energy conservation impose  $\sum_{i=1}^{\gamma-1} [n_a(\lambda_i)]/(\lambda_i) < [n_a(\lambda_\gamma)]/(\lambda_\gamma)$  with  $a = x, y, z$ .

## 1.7.3.2.2.1. Cubic crystals

There is no possibility of collinear phase matching in a dispersive cubic crystal because of the absence of birefringence. In a hypothetical non-dispersive anaxial crystal, the  $2^3$  three-wave and  $2^4$  four-wave phase-matching configurations would be allowed in any direction of propagation.

## 1.7.3.2.2.2. Uniaxial crystals

The configurations of polarization in terms of ordinary and extraordinary waves depend on the optic sign of the phase-matching direction with the convention given in Section 1.7.3.1: Tables 1.7.3.1 and 1.7.3.2 must be read by substituting (+, –) by (e, o) for a positive crystal and by (o, e) for a negative one.

Because of the symmetry of the index surface, all the phase-matching directions for a given type describe a cone with the optic axis as a revolution axis. Note that the previous comment on the anaxial class is valid for a propagation along the optic axis ( $n_o = n_e$ ).

Fig. 1.7.3.4 shows the example of negative uniaxial crystals ( $n_o > n_e$ ) like  $\beta\text{-BaB}_2\text{O}_4$  (BBO) and  $\text{KH}_2\text{PO}_4$  (KDP).

From Fig. 1.7.3.4, it clearly appears that the intersection of the sheets is possible only if  $(n_{e_\gamma})/(\lambda_\gamma) < \sum_{i=1}^{\gamma-1} (n_{o_i})/(\lambda_i) [< (n_{o_\gamma})/(\lambda_\gamma)]$  with  $\gamma = 3$  for a three-wave process and  $\gamma = 4$  for a four-wave one. The same considerations can be made for the positive sign and for all the other types of phase matching. There are different situations of inequalities allowing zero, one or several types: Table 1.7.3.3 gives the five possible situations for the three-wave interactions and Table 1.7.3.4 the 19 situations for the four-wave processes.

## 1.7.3.2.2.3. Biaxial crystals

The situation of biaxial crystals is more complicated, because the two sheets that must intersect are both elliptical in several cases. For a given interaction, all the phase-matching directions generate a complicated cone which joins two directions in the principal planes; the possible loci  $a, b, c, d$  are shown on the stereographic projection given in Fig. 1.7.3.5.

The basic inequalities of normal dispersion (1.7.3.7) forbid collinear phase matching for all the directions of propagation located between two optic axes at the two frequencies concerned.

Table 1.7.3.2. Correspondence between the phase-matching relations, the configurations of polarization and the types according to SFG ( $\omega_4 = \omega_1 + \omega_2 + \omega_3$ ), DFG ( $\omega_1 = \omega_4 - \omega_2 - \omega_3$ ), DFG ( $\omega_2 = \omega_4 - \omega_1 - \omega_3$ ) and DFG ( $\omega_3 = \omega_4 - \omega_1 - \omega_2$ ) (Boulanger et al., 1993)

Phase-matching relations	Configurations of polarization				Types of interaction			
	$\omega_4$	$\omega_1$	$\omega_2$	$\omega_3$	SFG ( $\omega_4$ )	DFG ( $\omega_1$ )	DFG ( $\omega_2$ )	DFG ( $\omega_3$ )
$\omega_4 n_4^+ = \omega_1 n_1^+ + \omega_2 n_2^+ + \omega_3 n_3^+$	e <sup>–</sup>	e <sup>+</sup>	e <sup>+</sup>	e <sup>+</sup>	I	II	III	IV
$\omega_4 n_4^- = \omega_1 n_1^- + \omega_2 n_2^- + \omega_3 n_3^-$	e <sup>–</sup>	e <sup>–</sup>	e <sup>–</sup>	e <sup>–</sup>	II	III	IV	I
$\omega_4 n_4^+ = \omega_1 n_1^- + \omega_2 n_2^+ + \omega_3 n_3^-$	e <sup>–</sup>	e <sup>–</sup>	e <sup>+</sup>	e <sup>–</sup>	III	IV	I	II
$\omega_4 n_4^- = \omega_1 n_1^+ + \omega_2 n_2^- + \omega_3 n_3^-$	e <sup>–</sup>	e <sup>+</sup>	e <sup>–</sup>	e <sup>–</sup>	IV	I	II	IV
$\omega_4 n_4^- = \omega_1 n_1^- + \omega_2 n_2^+ + \omega_3 n_3^+$	e <sup>–</sup>	e <sup>–</sup>	e <sup>+</sup>	e <sup>+</sup>	$\text{V}^4$	$\text{VI}^1$	$\text{V}^2$	$\text{V}^3$
$\omega_4 n_4^- = \omega_1 n_1^+ + \omega_2 n_2^- + \omega_3 n_3^+$	e <sup>–</sup>	e <sup>+</sup>	e <sup>–</sup>	e <sup>+</sup>	$\text{VI}^4$	$\text{VI}^1$	$\text{VI}^2$	$\text{VI}^3$
$\omega_4 n_4^- = \omega_1 n_1^+ + \omega_2 n_2^+ + \omega_3 n_3^-$	e <sup>–</sup>	e <sup>+</sup>	e <sup>+</sup>	e <sup>–</sup>	$\text{VII}^4$	$\text{VII}^1$	$\text{VII}^2$	$\text{VII}^3$

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Table 1.7.3.3. *Classes of refractive-index inequalities for collinear phase matching of three-wave interactions in positive and negative uniaxial crystals*

Types I, II and III refer to SFG; the types of the corresponding DFG are given in Table 1.7.3.1 (Fève *et al.*, 1993).

Positive sign ( $n_e > n_o$ )	Negative sign ( $n_o > n_e$ )	Types of SFG
$\frac{n_{o3}}{\lambda_3} < \frac{n_{o1}}{\lambda_1} + \frac{n_{e2}}{\lambda_2}, \frac{n_{e1}}{\lambda_1} + \frac{n_{o2}}{\lambda_2}$	$\frac{n_{o1}}{\lambda_1} + \frac{n_{e2}}{\lambda_2}, \frac{n_{e1}}{\lambda_1} + \frac{n_{o2}}{\lambda_2} < \frac{n_{e3}}{\lambda_3}$	I, II, III
$\frac{n_{e1}}{\lambda_1} + \frac{n_{o2}}{\lambda_2} < \frac{n_{o3}}{\lambda_3} < \frac{n_{o1}}{\lambda_1} + \frac{n_{e2}}{\lambda_2}$	$\frac{n_{o1}}{\lambda_1} + \frac{n_{e2}}{\lambda_2} < \frac{n_{e3}}{\lambda_3} < \frac{n_{e1}}{\lambda_1} + \frac{n_{o2}}{\lambda_2}$	I, II
$\frac{n_{o1}}{\lambda_1} + \frac{n_{e2}}{\lambda_2} < \frac{n_{o3}}{\lambda_3} < \frac{n_{e1}}{\lambda_1} + \frac{n_{o2}}{\lambda_2}$	$\frac{n_{e1}}{\lambda_1} + \frac{n_{o2}}{\lambda_2} < \frac{n_{e3}}{\lambda_3} < \frac{n_{o1}}{\lambda_1} + \frac{n_{e2}}{\lambda_2}$	I, III
$\frac{n_{o1}}{\lambda_1} + \frac{n_{e2}}{\lambda_2}, \frac{n_{e1}}{\lambda_1} + \frac{n_{o2}}{\lambda_2} < \frac{n_{o3}}{\lambda_3} < \frac{n_{e1}}{\lambda_1} + \frac{n_{e2}}{\lambda_2}$	$\frac{n_{o1}}{\lambda_1} + \frac{n_{e2}}{\lambda_2}, \frac{n_{e1}}{\lambda_1} + \frac{n_{o2}}{\lambda_2} < \frac{n_{e3}}{\lambda_3} < \frac{n_{o1}}{\lambda_1} + \frac{n_{o2}}{\lambda_2}$	I
$\frac{n_{e1}}{\lambda_1} + \frac{n_{e2}}{\lambda_2} < \frac{n_{o3}}{\lambda_3}$	$\frac{n_{o1}}{\lambda_1} + \frac{n_{o2}}{\lambda_2} < \frac{n_{e3}}{\lambda_3}$	None

Table 1.7.3.4. *Classes of refractive-index inequalities for collinear phase matching of four-wave interactions in positive ( $n_a = n_e, n_b = n_o$ ) and negative ( $n_a = n_o, n_b = n_e$ ) uniaxial crystals with  $(n_{b4}/\lambda_4) < (n_{a1}/\lambda_1) + (n_{a2}/\lambda_2) + (n_{a3}/\lambda_3)$*

If this inequality is not verified, no phase matching is allowed. The types of phase matching refer to SFG; the types of the corresponding DFG are given in Table 1.7.3.2 (Fève, 1994).

Positive sign ( $n_e > n_o$ )	Negative sign ( $n_o > n_e$ )	Types of SFG
$\frac{n_{a1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3}, \frac{n_{a1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3}, \frac{n_{b1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3} < \frac{n_{b4}}{\lambda_4}$		I
$\frac{n_{a1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3}, \frac{n_{a1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3} < \frac{n_{b4}}{\lambda_4} < \frac{n_{b1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3}$		I, V <sup>4</sup>
$\frac{n_{a1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3}, \frac{n_{b1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3} < \frac{n_{b4}}{\lambda_4} < \frac{n_{a1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3}$		I, VI <sup>4</sup>
$\frac{n_{a1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3}, \frac{n_{b1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3} < \frac{n_{b4}}{\lambda_4} < \frac{n_{a1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3}$		I, VII <sup>4</sup>
$\frac{n_{a1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3} < \frac{n_{b4}}{\lambda_4} < \frac{n_{a1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3}, \frac{n_{b1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3}$	$\frac{n_{b1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3} < \frac{n_{b4}}{\lambda_4}$	I, V <sup>4</sup> , VI <sup>4</sup>
	$\frac{n_{b4}}{\lambda_4} < \frac{n_{b1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3}$	I, II, V <sup>4</sup> , VI <sup>4</sup>
$\frac{n_{a1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3} < \frac{n_{b4}}{\lambda_4} < \frac{n_{a1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3}, \frac{n_{b1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3}$	$\frac{n_{b1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3} < \frac{n_{b4}}{\lambda_4}$	I, V <sup>4</sup> , VII <sup>4</sup>
	$\frac{n_{b4}}{\lambda_4} < \frac{n_{b1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3}$	I, III, V <sup>4</sup> , VII <sup>4</sup>
$\frac{n_{b1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3} < \frac{n_{b4}}{\lambda_4} < \frac{n_{a1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3}, \frac{n_{a1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3}$	$\frac{n_{a1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3} < \frac{n_{b4}}{\lambda_4}$	I, VI <sup>4</sup> , VII <sup>4</sup>
	$\frac{n_{b4}}{\lambda_4} < \frac{n_{a1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3}$	I, IV, VI <sup>4</sup> , VII <sup>4</sup>
$\frac{n_{b4}}{\lambda_4} < \frac{n_{a1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3}, \frac{n_{a1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3}, \frac{n_{b1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3}$	$\frac{n_{b1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3}, \frac{n_{b1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3}, \frac{n_{a1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3} < \frac{n_{b4}}{\lambda_4}$	I, V <sup>4</sup> , VI <sup>4</sup> , VII <sup>4</sup>
	$\frac{n_{a1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3}, \frac{n_{b1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3} < \frac{n_{b4}}{\lambda_4} < \frac{n_{b1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3}$	I, II, V <sup>4</sup> , VI <sup>4</sup> , VII <sup>4</sup>
	$\frac{n_{a1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3}, \frac{n_{b1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3} < \frac{n_{b4}}{\lambda_4} < \frac{n_{b1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3}$	I, III, V <sup>4</sup> , VI <sup>4</sup> , VII <sup>4</sup>
	$\frac{n_{b1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3}, \frac{n_{b1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3} < \frac{n_{b4}}{\lambda_4} < \frac{n_{a1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3}$	I, IV, V <sup>4</sup> , VI <sup>4</sup> , VII <sup>4</sup>
	$\frac{n_{a1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3} < \frac{n_{b4}}{\lambda_4} < \frac{n_{b1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3}, \frac{n_{b1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3}$	I, II, III, V <sup>4</sup> , VI <sup>4</sup> , VII <sup>4</sup>
	$\frac{n_{b1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3} < \frac{n_{b4}}{\lambda_4} < \frac{n_{a1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3}, \frac{n_{b1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3}$	I, II, IV, V <sup>4</sup> , VI <sup>4</sup> , VII <sup>4</sup>
	$\frac{n_{b1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3} < \frac{n_{b4}}{\lambda_4} < \frac{n_{a1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3}, \frac{n_{b1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3}$	I, III, IV, V <sup>4</sup> , VI <sup>4</sup> , VII <sup>4</sup>
	$\frac{n_{b4}}{\lambda_4} < \frac{n_{a1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3}, \frac{n_{b1}}{\lambda_1} + \frac{n_{a2}}{\lambda_2} + \frac{n_{b3}}{\lambda_3}, \frac{n_{b1}}{\lambda_1} + \frac{n_{b2}}{\lambda_2} + \frac{n_{a3}}{\lambda_3}$	All

# 1. TENSORIAL ASPECTS OF PHYSICAL PROPERTIES

Table 1.7.3.5. *Refractive-index conditions that determine collinear phase-matching loci in the principal planes of positive and negative biaxial crystals for three-wave SFG*

$a, b, c, d$  refer to the areas given in Fig. 1.7.3.5. The types corresponding to the different DFGs are given in Table 1.7.3.1 (Fève *et al.*, 1993).

Types of SFG	Phase-matching loci in the principal planes	Inequalities determining three-wave collinear phase matching in biaxial crystals	
		Positive biaxial crystal	Negative biaxial crystal
		$n_x(\omega_i) < n_y(\omega_i) < n_z(\omega_i)$	$n_x(\omega_i) > n_y(\omega_i) > n_z(\omega_i)$
Type I	$a$	$\frac{n_{x3}}{\lambda_3} < \frac{n_{y1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2} < \frac{n_{z3}}{\lambda_3}$	$\frac{n_{x1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2} > \frac{n_{y3}}{\lambda_3} > \frac{n_{z1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2}$
	$b$	$\frac{n_{x1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2} < \frac{n_{y3}}{\lambda_3} < \frac{n_{z1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2}$	$\frac{n_{x3}}{\lambda_3} > \frac{n_{y1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2} > \frac{n_{z3}}{\lambda_3}$
	$c$	$\frac{n_{x3}}{\lambda_3} < \frac{n_{z1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2} < \frac{n_{y3}}{\lambda_3}$	$\frac{n_{x1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2} > \frac{n_{z3}}{\lambda_3} > \frac{n_{y1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2}$
	$d$	$\frac{n_{y1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2} < \frac{n_{x3}}{\lambda_3} < \frac{n_{z1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2}$	$\frac{n_{y3}}{\lambda_3} > \frac{n_{x1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2} > \frac{n_{z3}}{\lambda_3}$
Type II	$a$	$\frac{n_{x3}}{\lambda_3} < \frac{n_{x1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2}; \frac{n_{z1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2} < \frac{n_{z3}}{\lambda_3}$	$\frac{n_{y1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2} > \frac{n_{y3}}{\lambda_3} > \frac{n_{y1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2}$
	$b$	$\frac{n_{y1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2} > \frac{n_{y3}}{\lambda_3} > \frac{n_{y1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2}$	$\frac{n_{x3}}{\lambda_3} > \frac{n_{x1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2}; \frac{n_{z1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2} > \frac{n_{z3}}{\lambda_3}$
	$c$	$\frac{n_{x3}}{\lambda_3} < \frac{n_{x1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2}; \frac{n_{y1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2} < \frac{n_{y3}}{\lambda_3}$	$\frac{n_{z1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2} > \frac{n_{z3}}{\lambda_3} > \frac{n_{z1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2}$
	$c^*$	$\frac{n_{x1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2} < \frac{n_{x3}}{\lambda_3}; \frac{n_{y3}}{\lambda_3} < \frac{n_{y1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2}$	$\frac{n_{z1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2} > \frac{n_{z3}}{\lambda_3} > \frac{n_{z1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2}$
	$d$	$\frac{n_{x1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2} < \frac{n_{x3}}{\lambda_3} < \frac{n_{x1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2}$	$\frac{n_{y3}}{\lambda_3} > \frac{n_{y1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2}; \frac{n_{z1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2} > \frac{n_{z3}}{\lambda_3}$
	$d^*$	$\frac{n_{x1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2} < \frac{n_{x3}}{\lambda_3} < \frac{n_{x1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2}$	$\frac{n_{y1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2} > \frac{n_{y3}}{\lambda_3}; \frac{n_{z3}}{\lambda_3} > \frac{n_{z1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2}$
Type III	$a$	$\frac{n_{x3}}{\lambda_3} < \frac{n_{y1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2}; \frac{n_{y1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2} < \frac{n_{z3}}{\lambda_3}$	$\frac{n_{x1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2} > \frac{n_{y3}}{\lambda_3} > \frac{n_{z1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2}$
	$b$	$\frac{n_{x1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2} < \frac{n_{y3}}{\lambda_3} < \frac{n_{z1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2}$	$\frac{n_{x3}}{\lambda_3} > \frac{n_{y1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2}; \frac{n_{y1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2} > \frac{n_{z3}}{\lambda_3}$
	$c$	$\frac{n_{x3}}{\lambda_3} < \frac{n_{z1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2}; \frac{n_{z1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2} < \frac{n_{y3}}{\lambda_3}$	$\frac{n_{x1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2} > \frac{n_{z3}}{\lambda_3} > \frac{n_{y1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2}$
	$c^*$	$\frac{n_{z1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2} < \frac{n_{x3}}{\lambda_3}; \frac{n_{y3}}{\lambda_3} < \frac{n_{z1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2}$	$\frac{n_{x1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2} > \frac{n_{z3}}{\lambda_3} > \frac{n_{y1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2}$
	$d$	$\frac{n_{y1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2} < \frac{n_{x3}}{\lambda_3} < \frac{n_{z1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2}$	$\frac{n_{y3}}{\lambda_3} > \frac{n_{x1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2}; \frac{n_{x1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2} > \frac{n_{z3}}{\lambda_3}$
	$d^*$	$\frac{n_{y1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2} < \frac{n_{x3}}{\lambda_3} < \frac{n_{z1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2}$	$\frac{n_{x1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2} > \frac{n_{y3}}{\lambda_3}; \frac{n_{z3}}{\lambda_3} > \frac{n_{x1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2}$
Conditions $c, d$ are applied if		$\frac{n_{y1}}{\lambda_1} - \frac{n_{x1}}{\lambda_1}, \frac{n_{y2}}{\lambda_2} - \frac{n_{x2}}{\lambda_2} < \frac{n_{y3}}{\lambda_3} - \frac{n_{x3}}{\lambda_3}$	$\frac{n_{y1}}{\lambda_1} - \frac{n_{z1}}{\lambda_1}, \frac{n_{y2}}{\lambda_2} - \frac{n_{z2}}{\lambda_2} < \frac{n_{y3}}{\lambda_3} - \frac{n_{z3}}{\lambda_3}$
Conditions $c^*, d^*$ are applied if		$\frac{n_{y3}}{\lambda_3} - \frac{n_{x3}}{\lambda_3} < \frac{n_{y1}}{\lambda_1} - \frac{n_{x1}}{\lambda_1}, \frac{n_{y2}}{\lambda_2} - \frac{n_{x2}}{\lambda_2}$	$\frac{n_{y3}}{\lambda_3} - \frac{n_{z3}}{\lambda_3} < \frac{n_{y1}}{\lambda_1} - \frac{n_{z1}}{\lambda_1}, \frac{n_{y2}}{\lambda_2} - \frac{n_{z2}}{\lambda_2}$

Tables 1.7.3.5 and 1.7.3.6 give, respectively, the inequalities that determine collinear phase matching in the principal planes for the three types of three-wave SFG and for the seven types of four-wave SFG.

The inequalities in Table 1.7.3.5 show that a phase-matching cone which would join the directions  $a$  and  $d$  is not possible for any type of interaction, because the corresponding inequalities have an opposite sense. It is the same for a hypothetical cone joining  $b$  and  $c$ .

The existence of type-II or type-III SFG phase matching imposes the existence of type I, because the inequalities relative to type I are always satisfied whenever type II or type III exists.

However, type I can exist even if type II or type III is not allowed. A type-I phase-matched SFG in area  $c$  forbids phase-matching directions in area  $b$  for type-II and type-III SFG. The exclusion is the same between  $d$  and  $a$ . The consideration of all the possible combinations of the inequalities of Table 1.7.3.5 leads to 84 possible classes of phase-matching cones for both positive and negative biaxial crystals (Fève *et al.*, 1993; Fève, 1994). There are 14 classes for second harmonic generation (SHG) which correspond to the degenerated case ( $\omega_1 = \omega_2$ ) (Hobden, 1967).

The coexistence of the different types of four-wave phase matching is limited as for the three-wave case: a cone joining  $a$  and  $d$  or  $b$  and  $c$  is impossible for type-I SFG. Type I in area  $d$

## 1.7. NONLINEAR OPTICAL PROPERTIES

Table 1.7.3.6. *Refractive-index conditions that determine collinear phase-matching loci in the principal planes of positive and negative biaxial crystals for four-wave SFG*

The types corresponding to the different DFGs are given in Table 1.7.3.2 (Boulanger *et al.*, 1993).

(a) SFG type I.

Phase-matching loci in the principal planes	Inequalities determining four-wave collinear phase matching in biaxial crystals	
	Positive sign	Negative sign
<i>a</i>	$\frac{n_{x4}}{\lambda_4} < \frac{n_{y1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2} + \frac{n_{y3}}{\lambda_3} < \frac{n_{z4}}{\lambda_4}$	$\frac{n_{z1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2} + \frac{n_{z3}}{\lambda_3} < \frac{n_{y4}}{\lambda_4} < \frac{n_{x1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2} + \frac{n_{x3}}{\lambda_3}$
<i>b</i>	$\frac{n_{x1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2} + \frac{n_{x3}}{\lambda_3} < \frac{n_{y4}}{\lambda_4} < \frac{n_{z1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2} + \frac{n_{z3}}{\lambda_3}$	$\frac{n_{z4}}{\lambda_4} < \frac{n_{y1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2} + \frac{n_{y3}}{\lambda_3} < \frac{n_{x4}}{\lambda_4}$
<i>c</i>	$\frac{n_{x4}}{\lambda_4} < \frac{n_{z1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2} + \frac{n_{z3}}{\lambda_3} < \frac{n_{y4}}{\lambda_4}$	$\frac{n_{y1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2} + \frac{n_{y3}}{\lambda_3} < \frac{n_{z4}}{\lambda_4} < \frac{n_{x1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2} + \frac{n_{x3}}{\lambda_3}$
<i>d</i>	$\frac{n_{y1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2} + \frac{n_{y3}}{\lambda_3} < \frac{n_{x4}}{\lambda_4} < \frac{n_{z1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2} + \frac{n_{z3}}{\lambda_3}$	$\frac{n_{z4}}{\lambda_4} < \frac{n_{x1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2} + \frac{n_{x3}}{\lambda_3} < \frac{n_{y4}}{\lambda_4}$

(b) SFG type II ( $i = 1, j = 2, k = 3$ ), SFG type III ( $i = 3, j = 1, k = 2$ ), SFG type IV ( $i = 2, j = 3, k = 1$ ).

Phase-matching loci in the principal planes	Inequalities determining four-wave collinear phase matching in biaxial crystals	
	Positive sign	Negative sign
<i>a</i>	$\frac{n_{x4}}{\lambda_4} < \frac{n_{xi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k}; \frac{n_{zi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k} < \frac{n_{z4}}{\lambda_4}$	$\frac{n_{yi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k} < \frac{n_{y4}}{\lambda_4} < \frac{n_{xi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k}$
<i>b</i>	$\frac{n_{yi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k} < \frac{n_{y4}}{\lambda_4} < \frac{n_{zi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k}$	$\frac{n_{z4}}{\lambda_4} < \frac{n_{zi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k}; \frac{n_{xi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k} < \frac{n_{x4}}{\lambda_4}$
<i>c</i>	$\frac{n_{x4}}{\lambda_4} < \frac{n_{xi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k}; \frac{n_{yi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k} < \frac{n_{y4}}{\lambda_4}$	$\frac{n_{zi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k} < \frac{n_{z4}}{\lambda_4} < \frac{n_{xi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k}$
<i>c*</i>	$\frac{n_{xi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k} < \frac{n_{x4}}{\lambda_4}; \frac{n_{y4}}{\lambda_4} < \frac{n_{yi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k}$	$\frac{n_{zi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k} < \frac{n_{z4}}{\lambda_4} < \frac{n_{xi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k}$
<i>d</i>	$\frac{n_{xi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k} < \frac{n_{x4}}{\lambda_4} < \frac{n_{zi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k}$	$\frac{n_{z4}}{\lambda_4} < \frac{n_{zi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k}; \frac{n_{yi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k} < \frac{n_{y4}}{\lambda_4}$
<i>d*</i>	$\frac{n_{xi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k} < \frac{n_{x4}}{\lambda_4} < \frac{n_{zi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k}$	$\frac{n_{zi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k} < \frac{n_{z4}}{\lambda_4}; \frac{n_{y4}}{\lambda_4} < \frac{n_{yi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k}$
SFG type II ( $i, j$ ) = (1, 2); SFG type III ( $i, j$ ) = (1, 3); SFG type IV ( $i, j$ ) = (2, 3)		
Conditions <i>c, d</i> are applied if	$\frac{n_{yi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} - \frac{n_{xi}}{\lambda_i} - \frac{n_{xj}}{\lambda_j} < \frac{n_{y4}}{\lambda_4} - \frac{n_{x4}}{\lambda_4}$	$\frac{n_{yi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} - \frac{n_{zi}}{\lambda_i} - \frac{n_{zj}}{\lambda_j} < \frac{n_{y4}}{\lambda_4} - \frac{n_{z4}}{\lambda_4}$
Conditions <i>c*, d*</i> are applied if	$\frac{n_{y4}}{\lambda_4} - \frac{n_{x4}}{\lambda_4} < \frac{n_{yi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} - \frac{n_{xi}}{\lambda_i} - \frac{n_{xj}}{\lambda_j}$	$\frac{n_{y4}}{\lambda_4} - \frac{n_{z4}}{\lambda_4} < \frac{n_{yi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} - \frac{n_{zi}}{\lambda_i} - \frac{n_{zj}}{\lambda_j}$

forbids the six other types in *a*. The same restriction exists between *c* and *b*. Types II, III, IV, V<sup>4</sup>, VI<sup>4</sup> and VII<sup>4</sup> cannot exist without type I; other restrictions concern the relations between types II, III, IV and types V<sup>4</sup>, VI<sup>4</sup>, VII<sup>4</sup> (Fève, 1994). The counting of the classes of four-wave phase-matching cones obtained from all the possible combinations of the inequalities of Table 1.7.3.6 is complex and it has not yet been done.

For reasons explained later, it can be interesting to consider a non-collinear interaction. In this case, the projection of the vectorial phase-matching relation (1.7.3.26) on the wavevector  $\mathbf{k}(\omega_\gamma, \theta_\gamma, \varphi_\gamma)$  of highest frequency  $\omega_\gamma$  leads to

$$\sum_{i=1}^{\gamma-1} \omega_i n(\omega_i, \theta_i, \varphi_i) \cos \alpha_{i\gamma} = \omega_\gamma n(\omega_\gamma, \theta_\gamma, \varphi_\gamma), \quad (1.7.3.29)$$

where  $\alpha_{i\gamma}$  is the angle between  $\mathbf{k}(\omega_i, \theta_i, \varphi_i)$  and  $\mathbf{k}(\omega_\gamma, \theta_\gamma, \varphi_\gamma)$ , with  $\gamma = 3$  for a three-wave interaction and  $\gamma = 4$  for a four-wave

interaction. The phase-matching angles ( $\theta_\gamma, \varphi_\gamma$ ) can be expressed as a function of the different ( $\theta_i, \varphi_i$ ) by the projection of (1.7.3.26) on the three principal axes of the optical frame.

The configurations of polarization allowing non-collinear phase matching are the same as for collinear phase matching. Furthermore, non-collinear phase matching exists only if collinear phase matching is allowed; the converse is not true (Fève, 1994). Note that collinear or non-collinear phase-matching conditions are rarely satisfied over the entire transparency range of the crystal.

### 1.7.3.2.3. Quasi phase matching

When index matching is not allowed, it is possible to increase the energy of the generated wave continuously during the propagation by introducing a periodic change in the sign of the nonlinear electric susceptibility, which leads to a periodic reset of