

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>  | $\frac{n_{yi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k} < \frac{n_{y4}}{\lambda_4} < \frac{n_{xi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k}$                           | $\frac{n_{z4}}{\lambda_4} < \frac{n_{zi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k}; \frac{n_{xi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k} < \frac{n_{x4}}{\lambda_4}$ |
| <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>  | $\frac{n_{xi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k} < \frac{n_{x4}}{\lambda_4} < \frac{n_{xi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k}$                           | $\frac{n_{z4}}{\lambda_4} < \frac{n_{zi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k}; \frac{n_{yi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k} < \frac{n_{y4}}{\lambda_4}$ |
| <i>d*</i>   | $\frac{n_{xi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k} < \frac{n_{x4}}{\lambda_4} < \frac{n_{xi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k}$                           | $\frac{n_{zi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k} < \frac{n_{z4}}{\lambda_4}; \frac{n_{y4}}{\lambda_4} < \frac{n_{yi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k}$ |
| SFG type II ( <i>i, j</i> ) = (1, 2); SFG type III ( <i>i, j</i> ) = (1, 3); SFG type IV ( <i>i, j</i> ) = (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 **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 **k**( $\omega_i, \theta_i, \varphi_i$ ) and **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

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Table 1.7.3.6 (cont.)

(c) SFG type  $V^4$  ( $i = 1, j = 2, k = 3$ ), SFG type  $VI^4$  ( $i = 2, j = 3, k = 1$ ), SFG type  $VII^4$  ( $i = 3, j = 1, k = 2$ ).

| Phase-matching loci in the principal planes   | Inequalities determining four-wave collinear phase matching in biaxial crystals  |  |
|---|--|--|
|   | Positive sign  | Negative sign  |
| $a$   | $\frac{n_{x4}}{\lambda_4} < \frac{n_{xi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k}; \frac{n_{zi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k} < \frac{n_{z4}}{\lambda_4}$ | $\frac{n_{yi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k} < \frac{n_{y4}}{\lambda_4} < \frac{n_{yi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k}$                           |
| $b$   | $\frac{n_{yi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k} < \frac{n_{y4}}{\lambda_4} < \frac{n_{yi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k}$                           | $\frac{n_{z4}}{\lambda_4} < \frac{n_{zi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k}; \frac{n_{xi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k} < \frac{n_{x4}}{\lambda_4}$ |
| $c'$  | $\frac{n_{x4}}{\lambda_4} < \frac{n_{xi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k}; \frac{n_{yi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k} < \frac{n_{y4}}{\lambda_4}$ | $\frac{n_{zi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k} < \frac{n_{z4}}{\lambda_4} < \frac{n_{zi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k}$                           |
| $c^{**}$  | $\frac{n_{xi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k} < \frac{n_{x4}}{\lambda_4}; \frac{n_{y4}}{\lambda_4} < \frac{n_{yi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k}$ | $\frac{n_{zi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k} < \frac{n_{z4}}{\lambda_4} < \frac{n_{zi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k}$                           |
| $d'$  | $\frac{n_{xi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k} < \frac{n_{x4}}{\lambda_4} < \frac{n_{xi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k}$                           | $\frac{n_{z4}}{\lambda_4} < \frac{n_{zi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k}; \frac{n_{yi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k} < \frac{n_{y4}}{\lambda_4}$ |
| $d^{**}$  | $\frac{n_{xi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k} < \frac{n_{x4}}{\lambda_4} < \frac{n_{xi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k}$                           | $\frac{n_{zi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k} < \frac{n_{z4}}{\lambda_4}; \frac{n_{y4}}{\lambda_4} < \frac{n_{yi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k}$ |
| SFG type $V^4$ ( $i = 1$ ); SFG type $VI^4$ ( $i = 2$ ); SFG type $VII^4$ ( $i = 3$ ) |  |  |
| Conditions $c', d'$ are applied if  | $\frac{n_{yi}}{\lambda_i} - \frac{n_{xi}}{\lambda_i} < \frac{n_{y4}}{\lambda_4} - \frac{n_{x4}}{\lambda_4}$  | $\frac{n_{yi}}{\lambda_i} - \frac{n_{zi}}{\lambda_i} < \frac{n_{y4}}{\lambda_4} - \frac{n_{z4}}{\lambda_4}$  |
| Conditions $c^{**}, d^{**}$ are applied if  | $\frac{n_{y4}}{\lambda_4} - \frac{n_{x4}}{\lambda_4} < \frac{n_{yi}}{\lambda_i} - \frac{n_{xi}}{\lambda_i}$  | $\frac{n_{y4}}{\lambda_4} - \frac{n_{z4}}{\lambda_4} < \frac{n_{yi}}{\lambda_i} - \frac{n_{zi}}{\lambda_i}$  |

$\pi$  between the waves (Armstrong *et al.*, 1962). This method is called quasi phase matching (QPM). The transfer of energy between the nonlinear polarization and the generated electric field never alternates if the reset is made at each coherence length. In this case and for a three-wave SFG, the nonlinear polarization sequence is the following:

(i) from 0 to  $L_c$ ,  $\mathbf{P}^{NL}(\omega_3) = \varepsilon_0 \chi^{(2)}(\omega_3) \mathbf{e}_1 \mathbf{e}_2 E_1 E_2 \exp\{i[k(\omega_1) + k(\omega_2)]Z\}$ ;

(ii) from  $L_c$  to  $2L_c$ ,  $\mathbf{P}^{NL}(\omega_3) = -\varepsilon_0 \chi^{(2)}(\omega_3) \mathbf{e}_1 \mathbf{e}_2 E_1 E_2 \exp\{i[k(\omega_1) + k(\omega_2)]Z\}$ , which is equivalent to  $\mathbf{P}^{NL}(\omega_3) = \varepsilon_0 \chi^{(2)}(\omega_3) \mathbf{e}_1 \mathbf{e}_2 E_1 E_2 \exp\{i[[k(\omega_1) + k(\omega_2)]Z - \pi]\}$ .

QPM devices are a recent development and are increasingly being considered for applications (Fejer *et al.*, 1992). The nonlinear medium can be formed by the bonding of thin wafers alternately rotated by  $\pi$ ; this has been done for GaAs (Gordon *et al.*, 1993). For ferroelectric crystals, it is possible to form periodic reversing of the spontaneous polarization in the same sample by proton- or ion-exchange techniques, or by applying an electric field, which leads to periodically poled (pp) materials like ppLiNbO<sub>3</sub> or ppKTiOPO<sub>4</sub> (Myers *et al.*, 1995; Karlsson & Laurell, 1997; Rosenman *et al.*, 1998).

Quasi phase matching offers three main advantages when compared with phase matching: it may be used for any configuration of polarization of the interacting waves, which allows us to use the largest coefficient of the  $\chi^{(2)}$  tensor, as explained in the following section; QPM can be achieved over the entire transparency range of the crystal, since the periodicity can be adjusted; and, finally, double refraction and its harmful effect on the nonlinear efficiency can be avoided because QPM can be realized in the principal plane of a uniaxial crystal or in the principal axes of biaxial crystals. Nevertheless, there are limitations due to the difficulty in fabricating the corresponding materials: diffusion-bonded GaAs has strong reflection losses and periodic patterns of ppKTP or ppLN can only be written over a thickness that does not exceed 3 mm, which limits the input energy.

## 1.7.3.2.4. Effective coefficient and field tensor

### 1.7.3.2.4.1. Definitions and symmetry properties

The refractive indices and their dispersion in frequency determine the existence and loci of the phase-matching directions, and so impose the direction of the unit electric field vectors of the interacting waves according to (1.7.3.9). The effective coefficient, given by (1.7.3.23) and (1.7.3.25), depends in part on the linear optical properties *via* the field tensor, which is the tensor product of the interacting unit electric field vectors (Boulanger, 1989; Boulanger & Marnier, 1991; Boulanger *et al.*, 1993; Zyss, 1993). Indeed, the effective coefficient is the contraction between the field tensor and the electric susceptibility tensor of corresponding order:

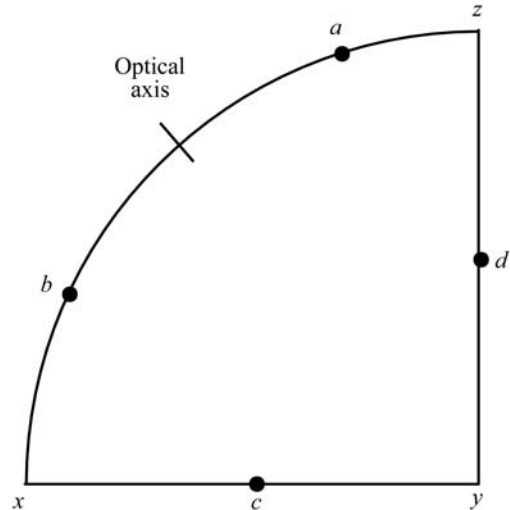


Fig. 1.7.3.5. Stereographic projection on the optical frame of the possible loci of phase-matching directions in the principal planes of a biaxial crystal.