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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	$\frac{n_{y4}}{\lambda_4}-\frac{n_{x4}}{\lambda_4} < \frac{n_{yi}}{\lambda_i}-\frac{n_{xi}}{\lambda_i}$	$\frac{n_{y4}}{\lambda_4}-\frac{n_{z4}}{\lambda_4} < \frac{n_{yi}}{\lambda_i}-\frac{n_{zi}}{\lambda_i}$

π 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, $\mathbf{P}^{NL}(\omega_3) = \varepsilon_0\chi^{(2)}(\omega_3)\mathbf{e}_1\mathbf{e}_2E_1E_2\exp\{i[k(\omega_1) + k(\omega_2)]Z\}$;

(ii) from L_c to $2L_c$, $\mathbf{P}^{NL}(\omega_3) = -\varepsilon_0\chi^{(2)}(\omega_3)\mathbf{e}_1\mathbf{e}_2E_1E_2\exp\{i[k(\omega_1) + k(\omega_2)]Z\}$, which is equivalent to $\mathbf{P}^{NL}(\omega_3) = \varepsilon_0\chi^{(2)}(\omega_3)\mathbf{e}_1\mathbf{e}_2E_1E_2\exp(i\{[k(\omega_1) + k(\omega_2)]Z - \pi\})$.

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 π; 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$_3$ or ppKTiOPO$_4$ (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:

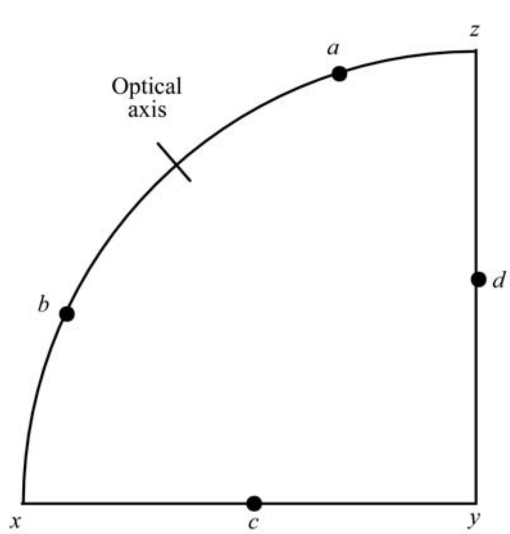

Fig. 1.7.3.5. Stereographic projection on the optical frame of the possible loci of phase-matching directions in the principal planes of a biaxial crystal.

(i) For three-wave mixing,

$$\chi^{(2)}_{\rm eff}(\omega_a, \omega_b, \omega_c, \theta, \varphi) = \sum_{ijk} \chi_{ijk}(\omega_a) F_{ijk}(\omega_a, \omega_b, \omega_c, \theta, \varphi) = \chi^{(2)}(\omega_a) \cdot F^{(2)}(\omega_a, \omega_b, \omega_c, \theta, \varphi), \quad (1.7.3.30)$$

with

$$F^{(2)}(\omega_a, \omega_b, \omega_c, \theta, \varphi) = \mathbf{e}(\omega_a, \theta, \varphi) \otimes \mathbf{e}(\omega_b, \theta, \varphi) \otimes \mathbf{e}(\omega_c, \theta, \varphi), \quad (1.7.3.31)$$

where $\omega_a, \omega_b, \omega_c$ correspond to $\omega_3, \omega_1, \omega_2$ for SFG ($\omega_3 = \omega_1 + \omega_2$); to $\omega_1, \omega_3, \omega_2$ for DFG ($\omega_1 = \omega_3 - \omega_2$); and to $\omega_2, \omega_3, \omega_1$ for DFG ($\omega_2 = \omega_3 - \omega_1$).

(ii) For four-wave mixing,

$$\chi^{(3)}_{\rm eff}(\omega_a, \omega_b, \omega_c, \omega_d, \theta, \varphi) = \sum_{ijkl} \chi_{ijkl}(\omega_a) F_{ijkl}(\omega_a, \omega_b, \omega_c, \omega_d, \theta, \varphi) = \chi^{(3)}(\omega_a) \cdot F^{(3)}(\omega_a, \omega_b, \omega_c, \omega_d, \theta, \varphi), \quad (1.7.3.32)$$

with

$$F^{(3)}(\omega_a, \omega_b, \omega_c, \omega_d \theta, \varphi) = \mathbf{e}(\omega_a, \theta, \varphi) \otimes \mathbf{e}(\omega_b, \theta, \varphi) \otimes \mathbf{e}(\omega_c, \theta, \varphi) \otimes \mathbf{e}(\omega_d, \theta, \varphi), \quad (1.7.3.33)$$

where $\omega_a, \omega_b, \omega_c, \omega_d$ correspond to $\omega_4, \omega_1, \omega_2, \omega_3$ for SFG ($\omega_4 = \omega_1 + \omega_2 + \omega_3$); to $\omega_1, \omega_4, \omega_2, \omega_3$ for DFG ($\omega_1 = \omega_4 - \omega_2 - \omega_3$); to $\omega_2, \omega_4, \omega_1, \omega_3$ for DFG ($\omega_2 = \omega_4 - \omega_1 - \omega_3$); and to ω_3, ω_4, ω_1, ω_2 for DFG ($\omega_3 = \omega_4 - \omega_1 - \omega_2$).

Each $\mathbf{e}(\omega_i, \theta, \varphi)$ corresponds to a given eigen electric field vector.

The components of the field tensor are trigonometric functions of the direction of propagation.

Particular relations exist between field-tensor components of SFG and DFG which are valid for any direction of propagation. Indeed, from (1.7.3.31) and (1.7.3.33), it is obvious that the field-tensor components remain unchanged by concomitant permutations of the electric field vectors at the different frequencies and the corresponding Cartesian indices (Boulanger & Marnier, 1991; Boulanger *et al.*, 1993):

$$F^{\mathbf{e}_3\mathbf{e}_1\mathbf{e}_2}_{ijk}(\omega_3 = \omega_1 + \omega_2) = F^{\mathbf{e}_1\mathbf{e}_3\mathbf{e}_2}_{jik}(\omega_1 = \omega_3 - \omega_2) = F^{\mathbf{e}_2\mathbf{e}_3\mathbf{e}_1}_{kij}(\omega_2 = \omega_3 - \omega_1) \quad (1.7.3.34)$$

and

$$\begin{aligned} &F^{\mathbf{e}_4\mathbf{e}_1\mathbf{e}_2\mathbf{e}_3}_{ijkl}(\omega_4 = \omega_1 + \omega_2 + \omega_3) \\ &= F^{\mathbf{e}_1\mathbf{e}_4\mathbf{e}_2\mathbf{e}_3}_{jikl}(\omega_1 = \omega_4 - \omega_2 - \omega_3) \\ &= F^{\mathbf{e}_2\mathbf{e}_4\mathbf{e}_1\mathbf{e}_3}_{kijl}(\omega_2 = \omega_4 - \omega_1 - \omega_3) \\ &= F^{\mathbf{e}_3\mathbf{e}_4\mathbf{e}_1\mathbf{e}_2}_{lijk}(\omega_3 = \omega_4 - \omega_1 - \omega_2), \end{aligned} \quad (1.7.3.35)$$

where $\mathbf{e}_i$ is the unit electric field vector at ω_i.

For a given interaction, the symmetry of the field tensor is governed by the vectorial properties of the electric fields, detailed in Section 1.7.3.1. This symmetry is then characteristic of both the optical class and the direction of propagation. These properties lead to four kinds of relations between the field-tensor components described later (Boulanger & Marnier, 1991; Boulanger *et al.*, 1993). Because of their interest for phase matching, we consider only the uniaxial and biaxial classes.

(*a*) The number of zero components varies with the direction of propagation according to the existence of nil electric field vector components. The only case where all the components are nonzero concerns any direction of propagation out of the principal planes in biaxial crystals.

(*b*) The orthogonality relation (1.7.3.10) between any ordinary and extraordinary waves propagating in the same direction leads to specific relations independent of the direction of propagation. For example, the field tensor of an (*eooo*) configuration of polarization (one extraordinary wave relative to the first Cartesian index and three ordinary waves relative to the three other indices) verifies $F_{xxij} + F_{yyij}$ $(+\ F_{zzij} = 0) =$ $F_{xixj} + F_{yiyj}$ $(+\ F_{zizj} = 0) = F_{xijx} + F_{yijy}$ $(+\ F_{zijz} = 0) = 0$, with i and j equal to x or y; the combination of these three relations leads to $F_{xxxx} = -F_{yyxx} = -F_{yxyx} = -F_{yxxy}, F_{yyyy} = -F_{xxyy} = -F_{xyxy} = -F_{xyyx}$ and $F_{yxyy} = F_{yyxy} = F_{yyyx} = -F_{xyxx} = F_{xxyx} = -F_{xxxy}$. In a biaxial crystal, this kind of relation does not exist out of the principal planes.

(*c*) The fact that the direction of the ordinary electric field vectors in uniaxial crystals does not depend on the frequency, (1.7.3.11), leads to symmetry in the Cartesian indices relative to the ordinary waves. These relations can be redundant in comparison with certain orthogonality relations and are valid for any direction of propagation in uniaxial crystals. It is also the case for biaxial crystals, but only in the principal planes xz and yz. In the xy plane of biaxial crystals, the ordinary wave, (1.7.3.15), has a walk-off angle which depends on the frequency, and the extraordinary wave, (1.7.3.16), has no walk-off angle: then the field tensor is symmetric in the Cartesian indices relative to the extraordinary waves. The walk-off angles of ordinary and extraordinary waves are nil along the principal axes of the index surface of biaxial and uniaxial crystals and so everywhere in the xy plane of uniaxial crystals. Thus, any field tensor associated with these directions of propagation is symmetric in the Cartesian indices relative to both the ordinary and extraordinary waves.

(*d*) Equalities between frequencies can create new symmetries: the field tensors of the uniaxial class for any direction of propagation and of the biaxial class in only the principal planes xz and yz become symmetric in the Cartesian indices relative to the extraordinary waves at the same frequency; in the xy plane of a biaxial crystal, this symmetry concerns the indices relative to the ordinary waves. Equalities between frequencies are the only situations for which the field tensors are partly symmetric out of the principal planes of a biaxial crystal: the symmetry concerns the indices relative to the waves (+) with identical frequencies; it is the same for the waves (−): for example, $F^{-++}_{ijk}(2\omega = \omega + \omega) = F^{-++}_{ikj}(2\omega = \omega + \omega)$, $F^{-++-}_{ijkl}(\omega_4 = \omega + \omega + \omega_3) = F^{-++-}_{ikjl}(\omega_4 = \omega + \omega + \omega_3)$, $F^{---+}_{ijkl}(\omega_4 = \omega + \omega + \omega_3) = F^{---+}_{ikjl}(\omega_4 = \omega + \omega + \omega_3)$ and so on.

1.7.3.2.4.2. *Uniaxial class*

The field-tensor components are calculated from (1.7.3.11) and (1.7.3.12). The phase-matching case is the only one considered here: according to Tables 1.7.3.1 and 1.7.3.2, the allowed configurations of polarization of three-wave and four-wave interactions, respectively, are the $2o.e$ (two ordinary and one extraordinary waves), the $2e.o$ and the $3o.e$, $3e.o$, $2o.2e$.

Tables 1.7.3.7 and 1.7.3.8 give, respectively, the matrix representations of the three-wave interactions (*eoo*), (*oee*) and of the four-wave (*oeee*), (*eooo*), (*ooee*) interactions for any direction of propagation in the general case where all the frequencies are different. In this situation, the number of independent components of the field tensors are: 7 for $2o.e$, 12 for $2e.o$, 9 for $3o.e$, 28 for $3e.o$ and 16 for $2o.2e$. Note that the increase of the number of ordinary waves leads to an enhancement of symmetry of the field tensors.

If there are equalities between frequencies, the field tensors *oee*, *oeee* and *ooee* become totally symmetric in the Cartesian indices relative to the extraordinary waves and the tensors *eoo* and *eooo* remain unchanged.

Table 1.7.3.9 gives the field-tensor components specifically nil in the principal planes of uniaxial and biaxial crystals. The nil

references