

## 1.7. NONLINEAR OPTICAL PROPERTIES

(i) For three-wave mixing,

$$\begin{aligned}\chi_{\text{eff}}^{(2)}(\omega_a, \omega_b, \omega_c, \theta, \varphi) &= \sum_{ijk} \chi_{ijk}(\omega_a) F_{ijk}(\omega_a, \omega_b, \omega_c, \theta, \varphi) \\ &= \chi^{(2)}(\omega_a) \cdot F^{(2)}(\omega_a, \omega_b, \omega_c, \theta, \varphi),\end{aligned}\quad (1.7.3.30)$$

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,

$$\begin{aligned}\chi_{\text{eff}}^{(3)}(\omega_a, \omega_b, \omega_c, \omega_d, \theta, \varphi) &= \sum_{ijkl} \chi_{ijkl}(\omega_a) F_{ijkl}(\omega_a, \omega_b, \omega_c, \omega_d, \theta, \varphi) \\ &= \chi^{(3)}(\omega_a) \cdot F^{(3)}(\omega_a, \omega_b, \omega_c, \omega_d, \theta, \varphi),\end{aligned}\quad (1.7.3.32)$$

with

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

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  $\omega_3, \omega_4, \omega_1, \omega_2$  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):

$$\begin{aligned}F_{ijk}^{\mathbf{e}_3 \mathbf{e}_1 \mathbf{e}_2}(\omega_3 = \omega_1 + \omega_2) &= F_{jik}^{\mathbf{e}_1 \mathbf{e}_3 \mathbf{e}_2}(\omega_1 = \omega_3 - \omega_2) \\ &= F_{kij}^{\mathbf{e}_2 \mathbf{e}_3 \mathbf{e}_1}(\omega_2 = \omega_3 - \omega_1)\end{aligned}\quad (1.7.3.34)$$

and

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

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

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 (*eo*) configuration of polarization (one extraordinary wave relative to the first Cartesian index and three ordinary waves relative to the three other indices) verifies  $F_{xxij} + F_{yyij} (+ F_{zzij} = 0) = F_{xixj} + F_{yiyj} (+ F_{zizj} = 0) = F_{xijx} + F_{yijy} (+ F_{zizj} = 0) = 0$ , with  $i$  and  $j$  equal to  $x$  or  $y$ ; the combination of these three relations leads to  $F_{xxxx} = -F_{yyxx} = -F_{yxyx} = -F_{yxyx}, F_{yyyy} = -F_{xxyy} = -F_{xyxy} = -F_{xyxy}$  and  $F_{xyyy} = F_{yyxy} = F_{yyxy} = -F_{yxxy} = F_{xxyx} = -F_{xxyx}$ . 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 (*eo*), (*oee*) and of the four-wave (*oeee*), (*eo*), (*oeee*) 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 *oeee* become totally symmetric in the Cartesian indices relative to the extraordinary waves and the tensors *eo* and *eo* 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

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components for the other configurations of polarization are obtained by permutation of the Cartesian indices and the corresponding polarizations.

From Tables 1.7.3.7 and 1.7.3.8, it is possible to deduce all the other  $2e.o$  interactions ( $eeo$ ), ( $oeo$ ), the  $2o.e$  interactions ( $ooe$ ), ( $oeo$ ), the  $3o.e$  interactions ( $oooe$ ), ( $oeoo$ ), ( $ooeo$ ), the  $3e.o$  interactions ( $eoee$ ), ( $eeoe$ ), ( $eeeo$ ) and the  $2o.2e$  interactions ( $oeoe$ ), ( $eeoo$ ), ( $oeeo$ ), ( $eeoe$ ). The corresponding interactions and types are given in Tables 1.7.3.1 and 1.7.3.2. According to (1.7.3.31) and (1.7.3.33), the magnitudes of two permuted components are equal if the permutation of polarizations are associated with the corresponding frequencies. For example, according to Table 1.7.3.2, two permuted field-tensor components have the same magnitude for permutation between the following  $3o.e$  interactions:

(i) ( $eoee$ ) SFG ( $\omega_4$ ) type I < 0 and the three ( $oeoo$ ) interactions, DFG ( $\omega_1$ ) type II < 0, DFG ( $\omega_2$ ) type III < 0, DFG ( $\omega_3$ ) type IV < 0;

(ii) the three ( $oooe$ ) interactions, SFG ( $\omega_4$ ) type II > 0, DFG ( $\omega_1$ ) type III > 0, DFG ( $\omega_2$ ) type IV > 0 and ( $eeoo$ ) DFG ( $\omega_3$ ) type I > 0;

(iii) the two ( $ooeo$ ) interactions SFG ( $\omega_4$ ) type III > 0, DFG ( $\omega_1$ ) type IV > 0, ( $eeoo$ ) DFG ( $\omega_2$ ) type I > 0, and ( $oooe$ ) DFG ( $\omega_3$ ) type II > 0;

(iv) ( $oeoo$ ) SFG ( $\omega_4$ ) type IV > 0, ( $eeoo$ ) DFG ( $\omega_1$ ) type I > 0, and the two interactions ( $ooeo$ ) DFG ( $\omega_2$ ) type II > 0, DFG ( $\omega_3$ ) type III > 0.

The contraction of the field tensor and the uniaxial dielectric susceptibility tensor of corresponding order, given in Tables 1.7.2.2 to 1.7.2.5, is nil for the following uniaxial crystal classes

Table 1.7.3.7. Matrix representations of the ( $oeo$ ) and ( $ooo$ ) field tensors of the uniaxial class and of the biaxial class in the principal planes  $xz$  and  $yz$ , with  $\omega_1 \neq \omega_2$  (Boulanger & Marnier, 1991)

$\bullet \quad F_{ijk} = 0$        $\bullet \text{---} \bullet \quad , \quad \circ \text{---} \circ \quad F_{ijk} = F_{lmn}$        $\bullet \text{---} \circ \quad F_{ijk} = -F_{lmn}$

Interactions	Three-rank $F_{ijk}(\theta, \varphi)$ field tensors
Type $ooo$ SFG ( $\omega_3$ ) type I < 0 DFG ( $\omega_1$ ) type I > 0 DFG ( $\omega_2$ ) type I > 0	
Type $oeo$ SFG ( $\omega_3$ ) type I > 0 DFG ( $\omega_1$ ) type I < 0 DFG ( $\omega_2$ ) type I < 0	

Table 1.7.3.8. Matrix representations of the ( $oeee$ ), ( $eoee$ ) and ( $ooee$ ) field tensors of the uniaxial class and of the biaxial class in the principal planes  $xz$  and  $yz$ , with  $\omega_1 \neq \omega_2 \neq \omega_3$  (Boulanger et al., 1993)

$\bullet \quad F_{ijkl} = 0$        $\bullet \text{---} \bullet \quad F_{ijkl} = F_{mnop}$        $\bullet \text{---} \circ \quad F_{ijkl} = -F_{mnop}$

Interactions	Four-rank $F_{ijkl}(\theta, \varphi)$ field tensors
Type $oeee$ SFG( $\omega_4$ ) type I > 0 DFG ( $\omega_1$ ) type I < 0 DFG ( $\omega_2$ ) type I < 0 DFG ( $\omega_3$ ) type I < 0	
Type $eoee$ SFG ( $\omega_4$ ) type I < 0 DFG ( $\omega_1$ ) type I > 0 DFG ( $\omega_2$ ) type I > 0 DFG ( $\omega_3$ ) type I > 0	
Type $ooee$ SFG ( $\omega_4$ ) type $V^4 > 0$ DFG ( $\omega_1$ ) type $V^1 > 0$ DFG ( $\omega_2$ ) type $V^2 > 0$ DFG ( $\omega_3$ ) type $V^3 > 0$	

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Table 1.7.3.9. Field-tensor components specifically nil in the principal planes of uniaxial and biaxial crystals for three-wave and four-wave interactions

( $i, j, k$ ) =  $x, y$  or  $z$ .

Configurations of polarization	Nil field-tensor components		
	(xy) plane	(xz) plane	(yz) plane
$ooo$	$F_{xjk} = 0; F_{yjk} = 0$	$F_{ixk} = F_{ijx} = 0$ $F_{yjk} = 0$	$F_{iyk} = F_{ijy} = 0$ $F_{xjk} = 0$
$oee$	$F_{ixk} = F_{ijx} = 0$ $F_{iyk} = F_{ijy} = 0$	$F_{iyk} = F_{ijy} = 0$ $F_{xik} = 0$	$F_{ixk} = F_{ijx} = 0$ $F_{yjk} = 0$
$oooo$	$F_{xjkl} = 0; F_{yjkl} = 0$	$F_{ixkl} = F_{ijxl} = F_{ijkx} = 0$ $F_{yjkl} = 0$	$F_{iykl} = F_{ijyl} = F_{ijkx} = 0$ $F_{xjkl} = 0$
$oeee$	$F_{ixkl} = F_{ijxl} = F_{ijkx} = 0$ $F_{iykl} = F_{ijyl} = F_{ijkx} = 0$	$F_{ixkl} = F_{ijxl} = F_{ijkx} = 0$ $F_{yjkl} = 0$	$F_{ixkl} = F_{ijxl} = F_{ijkx} = 0$ $F_{yjk} = 0$
$ooee$	$F_{ixkl} = F_{ijxl} = F_{ijkx} = 0$ $F_{iykl} = F_{ijyl} = F_{ijkx} = 0$	$F_{ixkl} = F_{ijxl} = F_{ijkx} = 0$ $F_{yjkl} = F_{ijyl} = F_{ijkx} = 0$	$F_{ixkl} = F_{ijxl} = F_{ijkx} = 0$ $F_{yjk} = F_{ijkx} = 0$

and configurations of polarization:  $D_4$  and  $D_6$  for  $2o.e$ ,  $C_{4v}$  and  $C_{6v}$  for  $2e.o$ ,  $D_6$ ,  $D_{6h}$ ,  $D_{3h}$  and  $C_{6v}$  for  $3o.e$  and  $3e.o$ . Thus, even if phase-matching directions exist, the effective coefficient in these situations is nil, which forbids the interactions considered (Boulanger & Marnier, 1991; Boulanger *et al.*, 1993). The number of forbidden crystal classes is greater under the Kleinman approximation. The forbidden crystal classes have been determined for the particular case of third harmonic generation assuming Kleinman conjecture and without consideration of the field tensor (Midwinter & Warner, 1965).

### 1.7.3.2.4.3. Biaxial class

The symmetry of the biaxial field tensors is the same as for the uniaxial class, though only for a propagation in the principal planes  $xz$  and  $yz$ ; the associated matrix representations are given in Tables 1.7.3.7 and 1.7.3.8, and the nil components are listed in Table 1.7.3.9. Because of the change of optic sign from either side of the optic axis, the field tensors of the interactions for which the phase-matching cone joins areas  $b$  and  $a$  or  $a$  and  $c$ , given in Fig. 1.7.3.5, change from one area to another: for example, the field tensor ( $oeoe$ ) becomes an ( $oooo$ ) and so the solicited components of the electric susceptibility tensor are not the same.

The nonzero field-tensor components for a propagation in the  $xy$  plane of a biaxial crystal are:  $F_{zxx}, F_{zyy}, F_{zxy} \neq F_{zyx}$  for ( $ooo$ );  $F_{xzz}, F_{yzz}$  for ( $oee$ );  $F_{zxxx}, F_{zyyy}, F_{zxyy} \neq F_{zyxy} \neq F_{zyyx}, F_{zxyx} \neq F_{zyyx} \neq F_{zyxx}$  for ( $oooo$ );  $F_{xzzz}, F_{yzzz}, F_{yzzz}$  for ( $oeee$ );  $F_{xyzz} \neq F_{yxzz}, F_{xxzz}, F_{yyzz}$  for ( $ooee$ ). The nonzero components for the other configurations of polarization are obtained by the associated permutations of the Cartesian indices and the corresponding polarizations.

The field tensors are not symmetric for a propagation out of the principal planes in the general case where all the frequencies are different: in this case there are 27 independent components for the three-wave interactions and 81 for the four-wave interactions, and so all the electric susceptibility tensor components are solicited.

As phase matching imposes the directions of the electric fields of the interacting waves, it also determines the field tensor and hence the effective coefficient. Thus there is no possibility of choice of the  $\chi^{(2)}$  coefficients, since a given type of phase matching is considered. In general, the largest coefficients of polar crystals, *i.e.*  $\chi_{zzz}$ , are implicated at a very low level when phase matching is achieved, because the corresponding field tensor, *i.e.*  $F_{zzz}$ , is often weak (Boulanger *et al.*, 1997). In contrast, QPM authorizes the coupling between three waves polarized along the  $z$  axis, which leads to an effective coefficient which is purely  $\chi_{zzz}$ , *i.e.*  $\chi_{\text{eff}} = (2/\pi)\chi_{zzz}$ , where the numerical factor comes from the periodic character of the rectangular function of modulation (Fejer *et al.*, 1992).

### 1.7.3.3. Integration of the propagation equations

#### 1.7.3.3.1. Spatial and temporal profiles

The resolution of the coupled equations (1.7.3.22) or (1.7.3.24) over the crystal length  $L$  leads to the electric field amplitude  $E_i(X, Y, L)$  of each interacting wave. The general solutions are Jacobian elliptic functions (Armstrong *et al.*, 1962; Fève, Boulanger & Douady, 2002). The integration of the systems is simplified for cases where one or several beams are held constant, which is called the undepleted pump approximation. We consider mainly this kind of situation here. The power of each interacting wave is calculated by integrating the intensity over the cross section of each beam according to (1.7.3.8). For our main purpose, we consider the simple case of plane-wave beams with two kinds of transverse profile:

$$\begin{aligned} \mathbf{E}(X, Y, Z) &= \mathbf{e}E_o(Z) \quad \text{for } (X, Y) \in [-w_o, +w_o] \\ \mathbf{E}(X, Y, Z) &= 0 \quad \text{elsewhere} \end{aligned} \quad (1.7.3.36)$$

for a flat distribution over a radius  $w_o$ ;

$$\mathbf{E}(X, Y, Z) = \mathbf{e}E_o(Z) \exp[-(X^2 + Y^2)/w_o^2] \quad (1.7.3.37)$$

for a Gaussian distribution, where  $w_o$  is the radius at  $(1/e)$  of the electric field and so at  $(1/e^2)$  of the intensity.

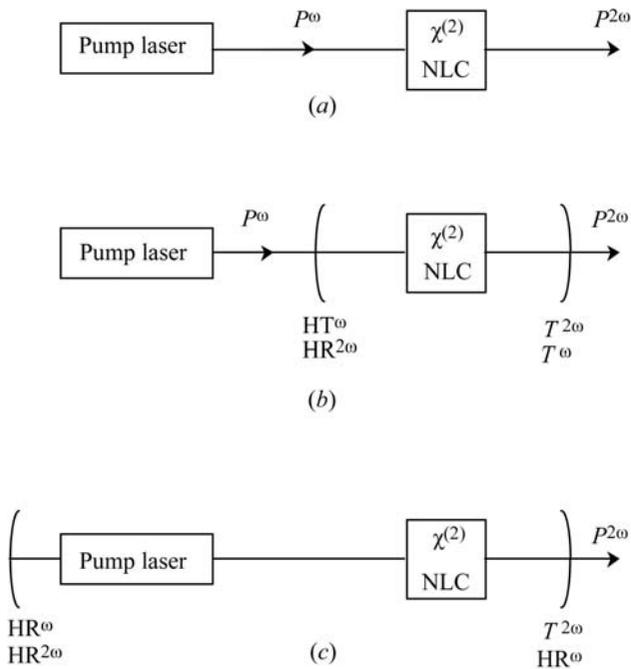


Fig. 1.7.3.6. Schematic configurations for second harmonic generation. (a) Non-resonant SHG; (b) external resonant SHG: the resonant wave may either be the fundamental or the harmonic one; (c) internal resonant SHG.  $P^{\omega, 2\omega}$  are the fundamental and harmonic powers;  $HT^{\omega}$  and  $HR^{\omega, 2\omega}$  are the high-transmission and high-reflection mirrors at  $\omega$  or  $2\omega$  and  $T^{\omega, 2\omega}$  are the transmission coefficients of the output mirror at  $\omega$  or  $2\omega$ . NLC is the nonlinear crystal with a nonzero  $\chi^{(2)}$ .