

## 1. TENSORIAL ASPECTS OF PHYSICAL PROPERTIES

(i) From the  $x$  axis to the optic axis,  $\mathbf{e}^o$  and  $\mathbf{e}^e$  are given by (1.7.3.11) and (1.7.3.12) with  $\varphi = 0$ . The walk-off is relative to the extraordinary wave and is calculated from (1.7.3.13) with  $n_o = n_x$  and  $n_e = n_z$ .

(ii) From the optic axis to the  $z$  axis, the vibration plane of the ordinary and extraordinary waves corresponds respectively to a rotation of  $\pi/2$  of the vibration plane of the extraordinary and ordinary waves for a propagation in the areas of the principal planes of opposite sign; the extraordinary electric field vector is given by (1.7.3.12) with  $\varphi = 0$ ,  $-\rho^-(\varphi, \omega)$  for the positive class and  $+\rho^+(\varphi, \omega)$  for the negative class, and the ordinary electric field vector is out of phase by  $\pi$  in relation to (1.7.3.11), that is

$$e_x^o = 0 \quad e_y^o = -1 \quad e_z^o = 0. \quad (1.7.3.17)$$

The extraordinary walk-off angle is given by (1.7.3.13) with  $n_o = n_x$  and  $n_e = n_z$ .

The  $\pi/2$  rotation on either side of the optic axes is well observed during internal conical refraction (Fève *et al.*, 1994).

Note that for a biaxial crystal, the walk-off angles are all nil only for a propagation along the principal axes.

## 1.7.3.1.4.2. Propagation out of the principal planes

It is impossible to define ordinary and extraordinary waves out of the principal planes of a biaxial crystal: according to (1.7.3.6) and (1.7.3.9),  $\mathbf{e}^+$  and  $\mathbf{e}^-$  have a nonzero projection on the  $z$  axis. According to these relations, it appears that  $\mathbf{e}^+$  and  $\mathbf{e}^-$  are not perpendicular, so relation (1.7.3.10) is never verified. The walk-off angles  $\rho^+$  and  $\rho^-$  are nonzero, different, and can be calculated from the electric field vectors:

$$\rho^\pm(\theta, \varphi, \omega) = \varepsilon \arccos[\mathbf{e}^\pm(\theta, \varphi, \omega) \cdot \mathbf{u}(\theta, \varphi, \omega)] - \varepsilon\pi/2. \quad (1.7.3.18)$$

$\varepsilon = +1$  or  $-1$  for a positive or a negative optic sign, respectively.

## 1.7.3.2. Equations of propagation of three-wave and four-wave interactions

## 1.7.3.2.1. Coupled electric fields amplitudes equations

The nonlinear crystals considered here are homogeneous, lossless, non-conducting, without optical activity, non-magnetic and are optically anisotropic. The nonlinear regime allows interactions between  $\gamma$  waves with different circular frequencies  $\omega_i$ ,  $i = 1, \dots, \gamma$ . The Fourier component of the polarization vector at  $\omega_i$  is  $\mathbf{P}(\omega_i) = \varepsilon_0 \chi^{(1)}(\omega_i) \mathbf{E}(\omega_i) + \mathbf{P}^{NL}(\omega_i)$ , where  $\mathbf{P}^{NL}(\omega_i)$  is the nonlinear polarization corresponding to the orders of the power series greater than 1 defined in Section 1.7.2.

Thus the propagation equation of each interacting wave  $\omega_i$  is (Bloembergen, 1965)

$$\nabla_x \nabla_x \mathbf{E}(\omega_i) = (\omega_i^2/c^2) \varepsilon(\omega_i) \mathbf{E}(\omega_i) + \omega_i^2 \mu_0 \mathbf{P}^{NL}(\omega_i). \quad (1.7.3.19)$$

The  $\gamma$  propagation equations are coupled by  $\mathbf{P}^{NL}(\omega_i)$ :

(1) for a three-wave interaction,  $\gamma = 3$ ,

$$\mathbf{P}^{NL}(\omega_1) = \mathbf{P}^{(2)}(\omega_1) = \varepsilon_0 \chi^{(2)}(\omega_1 = \omega_3 - \omega_2) \cdot \mathbf{E}(\omega_3) \otimes \mathbf{E}^*(\omega_2),$$

$$\mathbf{P}^{NL}(\omega_2) = \mathbf{P}^{(2)}(\omega_2) = \varepsilon_0 \chi^{(2)}(\omega_2 = \omega_3 - \omega_1) \cdot \mathbf{E}(\omega_3) \otimes \mathbf{E}^*(\omega_1),$$

$$\mathbf{P}^{NL}(\omega_3) = \mathbf{P}^{(2)}(\omega_3) = \varepsilon_0 \chi^{(2)}(\omega_3 = \omega_1 + \omega_2) \cdot \mathbf{E}(\omega_1) \otimes \mathbf{E}^*(\omega_2);$$

(2) for a four-wave interaction

$$\mathbf{P}^{NL}(\omega_1) = \mathbf{P}^{(3)}(\omega_1) = \varepsilon_0 \chi^{(3)}(\omega_1 = \omega_4 - \omega_2 - \omega_3) \cdot \mathbf{E}(\omega_4) \otimes \mathbf{E}^*(\omega_2) \otimes \mathbf{E}^*(\omega_3),$$

$$\mathbf{P}^{NL}(\omega_2) = \mathbf{P}^{(3)}(\omega_2) = \varepsilon_0 \chi^{(3)}(\omega_2 = \omega_4 - \omega_1 - \omega_3) \cdot \mathbf{E}(\omega_4) \otimes \mathbf{E}^*(\omega_1) \otimes \mathbf{E}^*(\omega_3),$$

$$\mathbf{P}^{NL}(\omega_3) = \mathbf{P}^{(3)}(\omega_3) = \varepsilon_0 \chi^{(3)}(\omega_3 = \omega_4 - \omega_1 - \omega_2) \cdot \mathbf{E}(\omega_4) \otimes \mathbf{E}^*(\omega_1) \otimes \mathbf{E}^*(\omega_2)$$

$$\mathbf{P}^{NL}(\omega_4) = \mathbf{P}^{(3)}(\omega_4) = \varepsilon_0 \chi^{(3)}(\omega_4 = \omega_1 + \omega_2 + \omega_3) \cdot \mathbf{E}(\omega_1) \otimes \mathbf{E}(\omega_2) \otimes \mathbf{E}(\omega_3).$$

The complex conjugates  $\mathbf{E}^*(\omega_i)$  come from the relation  $\mathbf{E}^*(\omega_i) = \mathbf{E}(-\omega_i)$ .

We consider the plane wave, (1.7.3.3), as a solution of (1.7.3.19), and we assume that all the interacting waves propagate in the same direction  $Z$ . Each linearly polarized plane wave corresponds to an eigen mode  $\mathbf{E}^+$  or  $\mathbf{E}^-$  defined above. For the usual case of beams with a finite transversal profile and when  $Z$  is along a direction where the double-refraction angles can be nonzero, *i.e.* out of the principal axes of the index surface, it is necessary to specify a frame for each interacting wave in order to calculate the corresponding powers as a function of  $Z$ : the coordinates linked to the wave at  $\omega_i$  are written  $(X_i, Y_i, Z)$ , which can be relative to the mode (+) or (-). The systems are then linked by the double-refraction angles  $\rho$ : according to Fig. 1.7.3.1, we have  $X_j^+ = X_i^+ + Z \tan[\rho^+(\omega_j) - \rho^+(\omega_i)]$ ,  $Y_j^+ = Y_i^+$  for two waves (+) with  $\rho^+(\omega_j) > \rho^+(\omega_i)$ , and  $X_j^- = X_i^-$ ,  $Y_j^- = Y_i^- + Z \tan[\rho^-(\omega_j) - \rho^-(\omega_i)]$  for two waves (-) with  $\rho^-(\omega_j) > \rho^-(\omega_i)$ .

The presence of  $\mathbf{P}^{NL}(\omega_i)$  in equations (1.7.3.19) leads to a variation of the  $\gamma$  amplitudes  $E(\omega_i)$  with  $Z$ . In order to establish the equations of evolution of the wave amplitudes, we assume that their variations are small over one wavelength  $\lambda_i$ , which is usually true. Thus we can state

$$\frac{1}{k(\omega_i)} \left| \frac{\partial E(\omega_i, X_i, Y_i, Z)}{\partial Z} \right| \ll |E(\omega_i, X_i, Y_i, Z)| \quad \text{or} \\ \left| \frac{\partial^2 E(\omega_i, X_i, Y_i, Z)}{\partial Z^2} \right| \ll k(\omega_i) \left| \frac{\partial E(\omega_i, X_i, Y_i, Z)}{\partial Z} \right|. \quad (1.7.3.20)$$

This is called the slowly varying envelope approximation.

Stating (1.7.3.20), the wave equation (1.7.3.19) for a forward propagation of a plane wave leads to

$$\frac{\partial E(\omega_i, X_i, Y_i, Z)}{\partial Z} = j\mu_0 \frac{\omega_i^2}{2k(\omega_i) \cos^2 \rho(\omega_i)} \mathbf{e}(\omega_i) \cdot \mathbf{P}^{NL}(\omega_i, X_i, Y_i, Z) \times \exp[-jk(\omega_i)Z]. \quad (1.7.3.21)$$

We choose the optical frame  $(x, y, z)$  for the calculation of all the scalar products  $\mathbf{e}(\omega_i) \cdot \mathbf{P}^{NL}(\omega_i)$ , the electric susceptibility tensors being known in this frame.

For a three-wave interaction, (1.7.3.21) leads to

$$\frac{\partial E_1(X_1, Y_1, Z)}{\partial Z} = j\kappa_1 [\mathbf{e}_1 \cdot \varepsilon_0 \chi^{(2)}(\omega_1 = \omega_3 - \omega_2) \cdot \mathbf{e}_3 \otimes \mathbf{e}_2] \times E_3(X_3, Y_3, Z) E_2^*(X_2, Y_2, Z) \exp(j\Delta k Z) \\ \frac{\partial E_2(X_2, Y_2, Z)}{\partial Z} = j\kappa_2 [\mathbf{e}_2 \cdot \varepsilon_0 \chi^{(2)}(\omega_2 = \omega_3 - \omega_1) \cdot \mathbf{e}_3 \otimes \mathbf{e}_1] \times E_3(X_3, Y_3, Z) E_1^*(X_1, Y_1, Z) \exp(j\Delta k Z) \\ \frac{\partial E_3(X_3, Y_3, Z)}{\partial Z} = j\kappa_3 [\mathbf{e}_3 \cdot \varepsilon_0 \chi^{(2)}(\omega_3 = \omega_1 + \omega_2) \cdot \mathbf{e}_1 \otimes \mathbf{e}_2] \times E_1(X_1, Y_1, Z) E_2(X_2, Y_2, Z) \exp(-j\Delta k Z), \quad (1.7.3.22)$$

with  $\mathbf{e}_i = \mathbf{e}(\omega_i)$ ,  $E_i(X_i, Y_i, Z) = E(\omega_i, X_i, Y_i, Z)$ ,  $\kappa_i = (\mu_0 \omega_i^2) / [2k(\omega_i) \cos^2 \rho(\omega_i)]$  and  $\Delta k = k(\omega_3) - [k(\omega_1) + k(\omega_2)]$ , called the phase mismatch. We take by convention  $\omega_1 < \omega_2 (< \omega_3)$ .

## 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:

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

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  $\Delta k Z + \Delta \Phi(Z) = \pi/2$ , 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 ( $\omega_2 = \omega_3 - \omega_1$ )

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

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