

## 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