(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 π in relation to (1.7.3.11), that is

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

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 ρ^+ and ρ^- are nonzero, different, and can be calculated from the electric field vectors:

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

$\varepsilon = +1$ 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 γ waves with different circular frequencies ω_i, $i = 1, \ldots, \gamma$. The Fourier component of the polarization vector at ω_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 ω_i is (Bloembergen, 1965)

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

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

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

$$\begin{aligned}
\mathbf{P}^{NL}(\omega_1) &= \mathbf{P}^{(2)}(\omega_1) = \varepsilon_0 \chi^{(2)}(\omega_1 = \omega_3 - \omega_2) \cdot \mathbf{E}(\omega_3) \otimes \mathbf{E}^*(\omega_2),\\
\mathbf{P}^{NL}(\omega_2) &= \mathbf{P}^{(2)}(\omega_2) = \varepsilon_0 \chi^{(2)}(\omega_2 = \omega_3 - \omega_1) \cdot \mathbf{E}(\omega_3) \otimes \mathbf{E}^*(\omega_1),\\
\mathbf{P}^{NL}(\omega_3) &= \mathbf{P}^{(2)}(\omega_3) = \varepsilon_0 \chi^{(2)}(\omega_3 = \omega_1 + \omega_2) \cdot \mathbf{E}(\omega_1) \otimes \mathbf{E}^*(\omega_2);
\end{aligned}$$

(2) for a four-wave interaction

$$\begin{aligned}
\mathbf{P}^{NL}(\omega_1) &= \mathbf{P}^{(3)}(\omega_1) = \varepsilon_0 \chi^{(3)}(\omega_1 = \omega_4 - \omega_2 - \omega_3)\\
&\qquad \cdot \mathbf{E}(\omega_4) \otimes \mathbf{E}^*(\omega_2) \otimes \mathbf{E}^*(\omega_3),\\
\mathbf{P}^{NL}(\omega_2) &= \mathbf{P}^{(3)}(\omega_2) = \varepsilon_0 \chi^{(3)}(\omega_2 = \omega_4 - \omega_1 - \omega_3)\\
&\qquad \cdot \mathbf{E}(\omega_4) \otimes \mathbf{E}^*(\omega_1) \otimes \mathbf{E}^*(\omega_3),\\
\mathbf{P}^{NL}(\omega_3) &= \mathbf{P}^{(3)}(\omega_3) = \varepsilon_0 \chi^{(3)}(\omega_3 = \omega_4 - \omega_1 - \omega_2)\\
&\qquad \cdot \mathbf{E}(\omega_4) \otimes \mathbf{E}^*(\omega_1) \otimes \mathbf{E}^*(\omega_2)\\
\mathbf{P}^{NL}(\omega_4) &= \mathbf{P}^{(3)}(\omega_4) = \varepsilon_0 \chi^{(3)}(\omega_4 = \omega_1 + \omega_2 + \omega_3)\\
&\qquad \cdot \mathbf{E}(\omega_1) \otimes \mathbf{E}(\omega_2) \otimes \mathbf{E}(\omega_3).
\end{aligned}$$

The complex conjugates $\mathbf{E}^*(\omega_1)$ 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 ω_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 ρ: 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 γ 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 λ_i, which is usually true. Thus we can state

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

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

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

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

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

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

references