International Tables for Crystallography (2013). Vol. D, Section 1.7.3.3.1, pp. 199–200.

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
eoo	$F_{xjk} = 0$; $F_{yjk} = 0$	$F_{ixk} = F_{ijx} = 0$ $F_{yjk} = 0$	$F_{iyk} = F_{ijy} = 0$ $F_{xjk} = 0$
oee	$F_{ixk} = F_{ijx} = 0$ $F_{iyk} = F_{ijy} = 0$	$F_{iyk} = F_{ijy} = 0$ $F_{xik} = 0$	$F_{ixk} = F_{ijx} = 0$ $F_{yjk} = 0$
eooo	$F_{xjkl} = 0$; $F_{yjkl} = 0$	$F_{ixkl} = F_{ijxl} = F_{ijkx} = 0$ $F_{yjkl} = 0$	$F_{iykl} = F_{ijyl} = F_{ijky} = 0$ $F_{xjkl} = 0$
oeee	$F_{ixkl} = F_{ijxl} = F_{ijkx} = 0$ $F_{iykl} = F_{ijyl} = F_{ijky} = 0$	$F_{iykl} = F_{ijyl} = F_{ijky} = 0$ $F_{xjkl} = 0$	$F_{ixkl} = F_{ijxl} = F_{ijkx} = 0$ $F_{yjkl} = 0$
ooee	$F_{ijxl} = F_{ijkx} = 0$ $F_{ijyl} = F_{ijky} = 0$	$F_{xjkl} = F_{ixkl} = 0$ $F_{ijyl} = F_{ijky} = 0$	$F_{yjkl} = F_{iykl} = 0$ $F_{ijxl} = F_{ijkx} = 0$

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 (*eoee*) becomes an (*oeoo*) and so the solicited components of the electric susceptibility tensor are not the same.

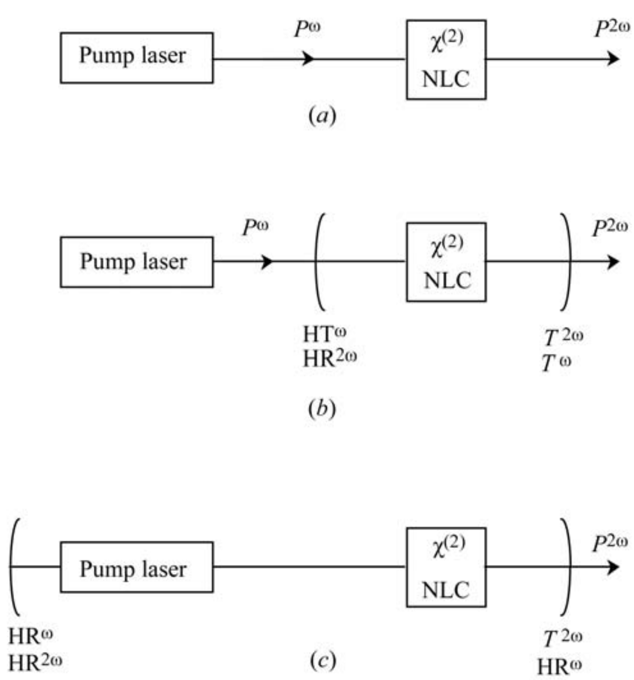

Fig. 1.7.3.6. Schematic configurations for second harmonic generation. (a) Non-resonant SHG; (b) external resonant SHG: the resonant wave may either be the fundamental or the harmonic one; (c) internal resonant SHG. $P^{\omega,2\omega}$ are the fundamental and harmonic powers; HT^{ω} and $\mathrm{HR}^{\omega,2\omega}$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

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 (*eoo*); F_{xzz}, F_{yzz} for (*oee*); F_{zxxx}, F_{zyyy}, $F_{zxyy} \neq F_{zyxy} \neq F_{zyyx}$, $F_{zxxy} \neq F_{zxyx} \neq F_{zyxx}$ for (*eooo*); F_{xzzz}, 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.* χ_{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 χ_{zzz}, *i.e.* $\chi_{\rm eff} = (2/\pi)\chi_{zzz}$, 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:

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

for a flat distribution over a radius w_o;

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

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.

The associated powers are calculated according to (1.7.3.8), which leads to

$$P(L) = m(n/2)(\varepsilon_o/\mu_o)^{1/2}|E_o|^2\pi w_o^2 \qquad (1.7.3.38)$$

where $m = 1$ for a flat distribution and $m = 1/2$ for a Gaussian profile.

The nonlinear interaction is characterized by the conversion efficiency, which is defined as the ratio of the generated power to the power of one or several incident beams, according to the different kinds of interactions.

For pulsed beams, it is necessary to consider the temporal shape, usually Gaussian:

$$P(t) = P_c \exp(-2t^2/\tau^2) \qquad (1.7.3.39)$$

where P_c is the peak power and τ the half $(1/e^2)$ width.

For a repetition rate f (s^{-1}), the average power $\tilde{P}$ is then given by

$$\tilde{P} = P_c\tau f(\pi/2)^{1/2} = \tilde{E}f \qquad (1.7.3.40)$$

where $\tilde{E}$ is the energy per Gaussian pulse.

When the pulse shape is not well defined, it is suitable to consider the energies per pulse of the incident and generated waves for the definition of the conversion efficiency.

The interactions studied here are sum-frequency generation (SFG), including second harmonic generation (SHG: $\omega + \omega = 2\omega$), cascading third harmonic generation (THG: $\omega + 2\omega = 3\omega$) and direct third harmonic generation (THG: $\omega + \omega + \omega = 3\omega$). The difference-frequency generation (DFG) is also considered, including optical parametric amplification (OPA) and oscillation (OPO).

We choose to analyse in detail the different parameters relative to conversion efficiency (figure of merit, acceptance bandwidths, walk-off effect *etc.*) for SHG, which is the prototypical second-order nonlinear interaction. This discussion will be valid for the other nonlinear processes of frequency generation which will be considered later.

1.7.3.3.2. *Second harmonic generation (SHG)*

According to Table 1.7.3.1, there are two types of phase matching for SHG: type I and type II (equivalent to type III).

The fundamental waves at ω define the pump. Two situations are classically distinguished: the undepleted pump approximation, when the power conversion efficiency is sufficiently low to consider the fundamental power to be undepleted, and the depleted case for higher efficiency. There are different ways to realize SHG, as shown in Fig. 1.7.3.6: the simplest one is non-resonant SHG, outside the laser cavity; other ways are external or internal resonant cavity SHG, which allow an enhancement of the single-pass efficiency conversion.

1.7.3.3.2.1. *Non-resonant SHG with undepleted pump in the parallel-beam limit with a Gaussian transverse profile*

We first consider the case where the crystal length is short enough to be located in the near-field region of the laser beam where the parallel-beam limit is a good approximation. We make another simplification by considering a propagation along a principal axis of the index surface: then the walk-off angle of each interacting wave is nil so that the three waves have the same coordinate system (X, Y, Z).

The integration of equations (1.7.3.22) over the crystal length Z in the undepleted pump approximation, *i.e.* $\partial E_1^{\omega}(X, Y, Z)/\partial Z = \partial E_2^{\omega}(X, Y, Z)/\partial Z = 0$, with $E_3^{2\omega}(X, Y, 0) = 0$, leads to

$$|E_3^{2\omega}(X, Y, L)|^2 = \{K_3^{2\omega}[\varepsilon_o\chi_{\rm eff}^{(2)}]\}^2 |E_1^{\omega}(X, Y, 0)E_2^{\omega}(X, Y, 0)|^2 \times L^2 \sin c^2[(\Delta k \cdot L)/2]. \qquad (1.7.3.41)$$

(1.7.3.41) implies a Gaussian transversal profile for $|E_3^{2\omega}(X, Y, L)|$ if $|E_1^{\omega}(X, Y, 0)|$ and $|E_2^{\omega}(X, Y, 0)|$ are Gaussian. The three beam radii are related by $(1/w_{o3}^2) = (1/w_{o1}^2) + (1/w_{o2}^2)$, so if we assume that the two fundamental beams have the same radius w_o^{ω}, which is not an approximation for type I, then $w_o^{2\omega} = [w_o^{\omega}/(2^{1/2})]$. Two incident beams with a flat distribution of radius w_o^{ω} lead to the generation of a flat harmonic beam with the same radius $w_o^{2\omega} = w_o^{\omega}$.

The integration of (1.7.3.41) according to (1.7.3.36)–(1.7.3.38) for a Gaussian profile gives in the SI system

$$P^{2\omega}(L) = BP_1^{\omega}(0)P_2^{\omega}(0)\frac{L^2}{w_o^2}\sin c^2\left(\frac{\Delta k \cdot L}{2}\right)$$
$$B = \frac{32\pi}{\varepsilon_o c}\frac{2N-1}{N}\frac{d_{\rm eff}^2}{\lambda_\omega^2}\frac{T_3^{2\omega}T_1^{\omega}T_2^{\omega}}{n_3^{2\omega}n_1^{\omega}n_2^{\omega}}, \quad (\mathrm{W}^{-1}) \qquad (1.7.3.42)$$

where $c = 3 \times 10^8$ m s^{-1}, $\varepsilon_o = 8.854 \times 10^{-12}$ A s V^{-1} m^{-1} and so $(32\pi/\varepsilon_o c) = 37.85 \times 10^3$ V A^{-1}. L (m) is the crystal length in the direction of propagation. $\Delta k = k_3^{2\omega} - k_1^{\omega} - k_2^{\omega}$ is the phase mismatch. $n_3^{2\omega}$, n_1^{ω} and n_2^{ω} are the refractive indices at the harmonic and fundamental wavelengths $\lambda_{2\omega}$ and λ_ω (µm): for the phase-matching case, $\Delta k = 0$, $n_3^{2\omega} = n^-(2\omega)$, $n_1^{\omega} = n_2^{\omega} = n^+(\omega)$ for type I (the two incident fundamental beams have the same polarization contained in Π^+, with the harmonic polarization contained in Π^-) and $n_1^{\omega} = n^+(\omega) \neq n_2^{\omega} = n^-(\omega)$ for type II (the two solicited eigen modes at the fundamental wavelength are in Π^+ and Π^-, with the harmonic polarization contained in Π^-). $T_3^{2\omega}$, T_1^{ω} and T_2^{ω} are the transmission coefficients given by $T_i = 4n_i/(n_i + 1)^2$. $d_{\rm eff}$ (pm V^{-1}) $= (1/2)\chi_{\rm eff} = (1/2)[F^{(2)} \cdot \chi^{(2)}]$ is the effective coefficient given by (1.7.3.30) and (1.7.3.31). $P_1^{\omega}(0)$ and $P_2^{\omega}(0)$ are the two incident fundamental powers, which are not necessarily equal for type II; for type I we have obviously $P_1^{\omega}(0) = P_2^{\omega}(0) = (P_{\rm tot}^{\omega}/2)$. N is the number of independently oscillating modes at the fundamental wavelength: every longitudinal mode at the harmonic pulsation can be generated by many combinations of two fundamental modes; the $(2N-1)/N$ factor takes into account the fluctuations between these longitudinal modes (Bloembergen, 1963).

The powers in (1.7.3.42) are instantaneous powers $P(t)$.

The second harmonic (SH) conversion efficiency, $\eta_{\rm SHG}$, is usually defined as the ratio of peak powers $P_c^{2\omega}(L)/P_{c,\rm tot}^{\omega}(0)$, or as the ratio of the pulse total energy $\tilde{E}^{2\omega}(L)/\tilde{E}_{\rm tot}^{\omega}(0)$. For Gaussian temporal profiles, the SH $(1/e^2)$ pulse duration $\tau_{2\omega}$ is equal to $\tau_\omega/(2^{1/2})$, because $P_{2\omega}$ is proportional to P_ω^2, and so, according to (1.7.3.40), the pulse average energy conversion efficiency is $1/(2^{1/2})$ smaller than the peak power conversion efficiency given by (1.7.3.42). Note that the pulse total energy conversion efficiency is equivalent to the average power conversion efficiency $\tilde{P}^{2\omega}(L)/\tilde{P}_{\rm tot}^{\omega}(0)$, with $\tilde{P} = \tilde{E} \cdot f$ where f is the repetition rate.

Formula (1.7.3.42) shows the importance of the contribution of the linear optical properties to the nonlinear process. Indeed, the field tensor $F^{(2)}$, the transmission coefficients T_i and the phase mismatch Δk only depend on the refractive indices in the direction of propagation considered.

(i) *Figure of merit*.

The contribution of $F^{(2)}$ was discussed previously, where it was shown that the field tensor is nil in particular directions of propagation or everywhere for particular crystal classes and configurations of polarization (even if the nonlinearity $\chi^{(2)}$ is high).

The field tensor $F^{(2)}$ of SHG can be written with the contracted notation of $d^{(2)}$; according to Table 1.7.3.1 and to the contraction