International Tables for Crystallography (2006). Vol. D, Section 1.7.3.3.4, pp. 207–208.

(n^ω, T^ω) are relative to the phase-matched SHG crystal and $(n_1^\omega, n_2^{2\omega}, n_3^{3\omega}, T_1^\omega, T_2^{2\omega}, T_3^{3\omega})$ correspond to the SFG crystal.

In the undepleted pump approximation for SHG, (1.7.3.71) becomes (Qiu & Penzkofer, 1988)

$$\eta_{\rm THG}(L_{\rm SHG}, L_{\rm SFG}) = BT^\omega \left[\frac{P^\omega(0)}{w_o^2}\right]^2 L_{\rm SHG}^2 L_{\rm SFG}^2 \sin c^2\left(\frac{\Delta k_{\rm SFG} L_{\rm SFG}}{2}\right) \quad (1.7.3.72)$$

with

$$B = B_{\rm SHG}\cdot B_{\rm SFG} = \frac{576\pi^2}{\varepsilon_o^2 c^2}\left(\frac{2N-1}{N}\right)^2 \frac{d^2_{{\rm eff}_{\rm SHG}} d^2_{{\rm eff}_{\rm SFG}}}{\lambda_\omega^4}\left(\frac{T^3_{\rm SHG}}{n^3_{\rm SHG}}\right)\left(\frac{T^3_{\rm SFG}}{n^3_{\rm SFG}}\right)$$

in W^{-2}, where

$$\frac{T^3_{\rm SHG}}{n^3_{\rm SHG}} = \frac{(T^\omega)^3}{(n^\omega)^3} \quad \text{and} \quad \frac{T^3_{\rm SFG}}{n^3_{\rm SFG}} = \frac{T_3^{3\omega} T_1^\omega T_2^{2\omega}}{n_3^{3\omega} n_1^\omega n_2^{2\omega}}.$$

The units are the same as in (1.7.3.42).

A more general case of SFG, where one of the two pump beams is depleted, is given in Section 1.7.3.3.4.

1.7.3.3.3.2. *SHG ($\omega + \omega = 2\omega$) and SFG ($\omega + 2\omega = 3\omega$) in the same crystal*

When the SFG conversion efficiency is sufficiently low in comparison with that of the SHG, it is possible to integrate the equations relative to SHG and those relative to SFG separately (Boulanger, Fejer *et al.*, 1994). In order to compare this situation with the example taken for the previous case, we consider a type-I configuration of polarization for SHG. By assuming a perfect phase matching for SHG, the amplitude of the third harmonic field inside the crystal is (Boulanger, 1994)

$$E^{3\omega}(X,Y,Z) = jK^{3\omega}(\varepsilon_o \chi_{{\rm eff}_{\rm SFG}}) \times \int_0^L E^\omega_{\rm tot}(X,Y,Z) E^{2\omega}(X,Y,Z)\exp(j\Delta k_{\rm SFG} Z)\,{\rm d}Z \quad (1.7.3.73)$$

with

$$E^{2\omega}(X,Y,Z) = (T^\omega)^{1/2}|E^\omega_{\rm tot}(0)|\tanh(\Gamma Z)$$
$$\text{and} \quad E^\omega_{\rm tot}(X,Y,Z) = (T^\omega)^{1/2}|E^\omega_{\rm tot}(0)|\,{\rm sech}(\Gamma Z). \quad (1.7.3.74)$$

Γ is as in (1.7.3.59).

(1.7.3.73) can be analytically integrated for undepleted pump SHG; sech$(m) \to 1$, tanh$(m) \to m$, and so we have

$$\eta_{\rm THG}(L) = P^{3\omega}(L)/P^\omega_{\rm tot}(0) \quad (1.7.3.75)$$

with

$$P^{3\omega}(L) = \frac{576\pi^2}{\varepsilon_o^2 c^2}\left(\frac{2N-1}{N}\right)^2 T^{3\omega} \frac{d^2_{{\rm eff}_{\rm SHG}} d^2_{{\rm eff}_{\rm SFG}}}{n^{3\omega}(n^\omega)^3 (n^{2\omega})^2} \frac{[T^\omega P^\omega_{\rm tot}(0)]^3}{w_o^4 \lambda_\omega^4} J(L),$$

where the integral $J(L)$ is

$$J(L) = \left|\int_0^L Z\exp(i\Delta k_{\rm SFG} Z)\,{\rm d}Z\right|^2. \quad (1.7.3.76)$$

For a nonzero SFG phase mismatch, $\Delta k_{\rm SFG} \neq 0$,

$$J(L) \simeq L^2/(\Delta k_{\rm SFG})^2. \quad (1.7.3.77)$$

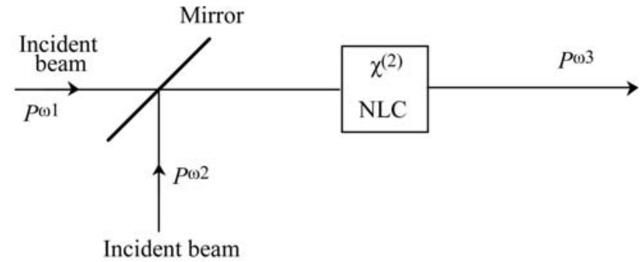

Fig. 1.7.3.17. Frequency up-conversion process $\omega_1 + \omega_2 = \omega_3$. The beam at ω_1 is mixed with the beam at ω_2 in the nonlinear crystal NLC in order to generate a beam at ω_3. $P^{\omega_1,\omega_2,\omega_3}$ are the different powers.

For phase-matched SFG, $\Delta k_{\rm SFG} = 0$,

$$J(L) = L^4/4. \quad (1.7.3.78)$$

Therefore (1.7.3.75) according to (1.7.3.78) is equal to (1.7.3.72) with $L_{\rm SHG} = L_{\rm SFG} = L/2$, $\Delta k_{\rm SFG} = 0$ and 100% transmission coefficients at ω and 2ω between the two crystals.

1.7.3.3.3.3. *Direct THG ($\omega + \omega + \omega = 3\omega$)*

As for the cascading process, we consider a flat plane wave which propagates in a direction without walk-off. The integration of equations (1.7.3.24) over the crystal length L, with $E_4^{3\omega}(X,Y,0) = 0$ and in the undepleted pump approximation, leads to

$$E_4^{3\omega}(X,Y,L) = jK_4^{3\omega}[\varepsilon_o \chi^{(3)}_{\rm eff}]E_1^\omega(X,Y,0)E_2^\omega(X,Y,0)E_3^\omega(X,Y,0) \times L\sin c[(\Delta k\cdot L)/2]\exp(-j\Delta kL/2). \quad (1.7.3.79)$$

According to (1.7.3.36) and (1.7.3.38), the integration of (1.7.3.79) over the cross section, which is the same for the four beams, leads to

$$\eta_{\rm THG}(L) = \frac{P^{3\omega}(L)}{P^\omega(0)} = B_{\rm THG}[P^\omega(0)]^2 \frac{L^2}{w_o^4}\sin c^2[(\Delta k\cdot L)/2]$$

with

$$B_{\rm THG} = \frac{576}{\varepsilon_o^2 c^2}\frac{d^2_{\rm eff}}{\lambda_\omega^2}\frac{T_4^{3\omega}(T_1^\omega)^2 T_2^\omega}{n_4^{3\omega}(n_1^\omega)^2 n_2^\omega} \quad ({\rm m}^2\ {\rm W}^{-2}), \quad (1.7.3.80)$$

where $d_{\rm eff} = (1/4)\chi^{(3)}_{\rm eff}$ is in $m^2 V^{-2}$ and λ_ω is in m. The statistical factor is assumed to be equal to 1, which corresponds to a longitudinal single-mode laser.

The different types of phase matching and the associated relations and configurations of polarization are given in Table 1.7.3.2 by considering the SFG case with $\omega_1 = \omega_2 = \omega_3 = \omega_4/3$.

1.7.3.3.4. *Sum-frequency generation (SFG)*

SHG ($\omega + \omega = 2\omega$) and SFG ($\omega + 2\omega = 3\omega$) are particular cases of three-wave SFG. We consider here the general situation where the two incident beams at ω_1 and ω_2, with $\omega_1 < \omega_2$, interact with the generated beam at ω_3, with $\omega_3 = \omega_1 + \omega_2$, as shown in Fig. 1.7.3.17. The phase-matching configurations are given in Table 1.7.3.1.

From the general point of view, SFG is a frequency up-conversion parametric process which is used for the conversion of laser beams at low circular frequency: for example, conversion of infrared to visible radiation.

The resolution of system (1.7.3.22) leads to Jacobian elliptic functions if the waves at ω_1 and ω_2 are both depleted. The calculation is simplified in two particular situations which are often encountered: on the one hand undepletion for the waves at ω_1 and ω_2, and on the other hand depletion of only one wave at

ω_1 or ω_2. For the following, we consider plane waves which propagate in a direction without walk-off so we consider a single wave frame; the energy distribution is assumed to be flat, so the three beams have the same radius w_o.

1.7.3.3.4.1. *SFG ($\omega_1 + \omega_2 = \omega_3$) with undepletion at ω_1 and ω_2*

The resolution of system (1.7.3.22) with $E_1(X, Y, 0) \neq 0$, $E_2(X, Y, 0) \neq 0$, $\partial E_1(X, Y, Z)/\partial Z = \partial E_2(X, Y, Z)/\partial Z = 0$ and $E_3(X, Y, 0) = 0$, followed by integration over (X, Y), leads to

$$P^{\omega_1}(L) = (T^{\omega_1})^2 P^{\omega_1}(0) \quad (1.7.3.81)$$

$$P^{\omega_2}(L) = (T^{\omega_2})^2 P^{\omega_2}(0) \quad (1.7.3.82)$$

$$P^{\omega_3}(L) = BP^{\omega_1}(0)P^{\omega_2}(0)\frac{L^2}{w_o^2}\sin c^2\frac{\Delta k \cdot L}{2} \quad (1.7.3.83)$$

with

$$B_{\rm SFG} = \frac{72\pi}{\varepsilon_o c}\frac{2N-1}{N}\frac{d_{\rm eff}^2}{\lambda_\omega^2}\frac{T^{\omega_3}T^{\omega_1}T^{\omega_2}}{n^{\omega_3}n^{\omega_1}n^{\omega_2}} \quad ({\rm W}^{-1})$$

in the same units as equation (1.7.3.70).

1.7.3.3.4.2. *SFG ($\omega_s + \omega_p = \omega_i$) with undepletion at ω_p*

$(\omega_s, \omega_p, \omega_i) = (\omega_1, \omega_2, \omega_3)$ or $(\omega_2, \omega_1, \omega_3)$.

The undepleted wave at ω_p, the pump, is mixed in the nonlinear crystal with the depleted wave at ω_s, the signal, in order to generate the idler wave at $\omega_i = \omega_s + \omega_p$. The integrations of the coupled amplitude equations over (X, Y, Z) with $E_s(X, Y, 0) \neq 0$, $E_p(X, Y, 0) \neq 0$, $\partial E_p(X, Y, Z)/\partial Z = 0$ and $E_i(X, Y, 0) = 0$ give

$$P_p(L) = T_p^2 P_p(0) \quad (1.7.3.84)$$

$$P_i(L) = \frac{\omega_i}{\omega_s}P_s(0)\Gamma^2L^2\frac{\sin^2\{\Gamma^2L^2 + [(\Delta k \cdot L)/2]^2\}^{1/2}}{\Gamma^2L^2 + [(\Delta k \cdot L)/2]^2} \quad (1.7.3.85)$$

$$P_s(L) = P_s(0)\left[1 - \frac{\omega_s}{\omega_i}\frac{P_i(L)}{P_s(0)}\right], \quad (1.7.3.86)$$

with $\Delta k = k_i - (k_s + k_p)$ and $\Gamma^2 = [B_s P_p(0)]/w_o^2$, where

$$B_s = \frac{8\pi}{\varepsilon_o c}\frac{2N-1}{N}\frac{d_{\rm eff}^2}{\lambda_s\lambda_i}\frac{T_sT_pT_i}{n_sn_pn_i}.$$

Thus, even if the up-conversion process is phase-matched ($\Delta k = 0$), the power transfers are periodic: the photon transfer efficiency is then 100% for $\Gamma L = (2m+1)(\pi/2)$, where m is an integer, which allows a maximum power gain ω_i/ω_s for the idler. A nonlinear crystal with length $L = (\pi/2\Gamma)$ is sufficient for an optimized device.

For a small conversion efficiency, *i.e.* ΓL weak, (1.7.3.85) and (1.7.3.86) become

$$P_i(L) \simeq P_s(0)\frac{\omega_i}{\omega_s}\Gamma^2L^2\sin c^2\frac{\Delta k \cdot L}{2} \quad (1.7.3.87)$$

and

$$P_s(L) \simeq P_s(0). \quad (1.7.3.88)$$

The expression for $P_i(L)$ with $\Delta k = 0$ is then equivalent to (1.7.3.83) with $\omega_p = \omega_1$ or ω_2, $\omega_i = \omega_3$ and $\omega_s = \omega_2$ or ω_1.

For example, the frequency up-conversion interaction can be of great interest for the detection of a signal, ω_s, comprising IR radiation with a strong divergence and a wide spectral bandwidth. In this case, the achievement of a good conversion efficiency, $P_i(L)/P_s(0)$, requires both wide spectral and angular acceptance bandwidths with respect to the signal. The double non-criticality in frequency and angle (DNPM) can then be used with one-beam non-critical non-collinear phase matching (OBNC) associated with vectorial group phase matching (VGPM) (Dolinchuk *et al.*, 1994): this corresponds to the equality of the absolute magnitudes and directions of the signal and idler group velocity vectors *i.e.* $d\omega_i/d\mathbf{k}_i = d\omega_s/d\mathbf{k}_s$.

1.7.3.3.5. *Difference-frequency generation (DFG)*

DFG is defined by $\omega_3 - \omega_1 = \omega_2$ with $E_2(X, Y, 0) = 0$ or $\omega_3 - \omega_2 = \omega_1$ with $E_1(X, Y, 0) = 0$. The DFG phase-matching configurations are given in Table 1.7.3.1. As for SFG, the solutions of system (1.7.3.22) are Jacobian elliptic functions when the incident waves are both depleted. We consider here the simplified situations of undepletion of the two incident waves and depletion of only one incident wave. In the latter, the solutions differ according to whether the circular frequency of the undepleted wave is the highest one, *i.e.* ω_3, or not. We consider the case of plane waves that propagate in a direction without walk-off and we assume a flat energy distribution for the three beams.

1.7.3.3.5.1. *DFG ($\omega_p - \omega_s = \omega_i$) with undepletion at ω_p and ω_s*

$(\omega_s, \omega_i, \omega_p) = (\omega_1, \omega_2, \omega_3)$ or $(\omega_2, \omega_1, \omega_3)$.

The resolution of system (1.7.3.22) with $E_s(X, Y, 0) \neq 0$, $E_p(X, Y, 0) \neq 0$, $\partial E_p(X, Y, Z)/\partial Z = \partial E_s(X, Y, Z)/\partial Z = 0$ and $E_i(X, Y, 0) = 0$, followed by integration over (X, Y), leads to the same solutions as for SFG with undepletion at ω_1 and ω_2, *i.e.* formulae (1.7.3.81), (1.7.3.82) and (1.7.3.83), by replacing ω_1 by ω_s, ω_2 by ω_p and ω_3 by ω_i. A schematic device is given in Fig. 1.7.3.17 by replacing $(\omega_1, \omega_2, \omega_3)$ by $(\omega_1, \omega_3, \omega_2)$ or $(\omega_2, \omega_3, \omega_1)$.

1.7.3.3.5.2. *DFG ($\omega_s - \omega_p = \omega_i$) with undepletion at ω_p*

$(\omega_s, \omega_i, \omega_p) = (\omega_3, \omega_1, \omega_2)$ or $(\omega_3, \omega_2, \omega_1)$.

The resolution of system (1.7.3.22) with $E_s(X, Y, 0) \neq 0$, $E_p(X, Y, 0) \neq 0$, $\partial E_p(X, Y, Z)/\partial Z = 0$ and $E_i(X, Y, 0) = 0$, followed by the integration over (X, Y), leads to the same solutions as for SFG with undepletion at ω_1 or ω_2: formulae (1.7.3.84), (1.7.3.85) and (1.7.3.86).

1.7.3.3.5.3. *DFG ($\omega_p - \omega_s = \omega_i$) with undepletion at ω_p – optical parametric amplification (OPA), optical parametric oscillation (OPO)*

$(\omega_s, \omega_i, \omega_p) = (\omega_1, \omega_2, \omega_3)$ or $(\omega_2, \omega_1, \omega_3)$.

The initial conditions are the same as in Section 1.7.3.3.5.2, except that the undepleted wave has the highest circular frequency. In this case, the integrations of the coupled amplitude equations over (X, Y, Z) lead to

$$P_p(L) = T_p^2 P_p(0), \quad (1.7.3.89)$$

$$P_i(L) = P_s(0)\frac{\omega_i}{\omega_s}\Gamma^2L^2\frac{\sinh^2\{\Gamma^2L^2 - [(\Delta k \cdot L)/2]^2\}^{1/2}}{\Gamma^2L^2 - [(\Delta k \cdot L)/2]^2} \quad (1.7.3.90)$$

and

$$P_s(L) = P_s(0)\left[1 + \frac{\omega_s}{\omega_i}\frac{P_i(L)}{P_s(0)}\right] = P_s(0)\left(1 + \Gamma^2L^2\frac{\sinh^2\{\Gamma^2L^2 - [(\Delta k \cdot L)/2]^2\}^{1/2}}{\Gamma^2L^2 - [(\Delta k \cdot L)/2]^2}\right) \quad (1.7.3.91)$$

with $\Delta k = k_p - (k_i + k_s)$ and $\Gamma^2 = [B_i P_p(0)]/w_o^2$, where w_o is the beam radius of the three beams and

$$B_i = \frac{8\pi}{\varepsilon_o c}\frac{2N-1}{N}\frac{d_{\rm eff}^2}{\lambda_s\lambda_i}\frac{T_sT_pT_i}{n_sn_pn_i}.$$