International Tables for Crystallography (2013). Vol. D, Section 1.7.3.3.3, pp. 209–210.

The intracavity SHG conversion efficiency is usually defined as the ratio of the SH output power to the maximum output power that would be obtained from the laser without the nonlinear crystal by optimal linear output coupling.

Maximizing (1.7.3.67) with respect to K according to (1.7.3.62), (1.7.3.65) and (1.7.3.66) gives (Perkins & Fahlen, 1987)

$$K_{\rm opt} = (\gamma_L + \gamma_{NL})S \qquad (1.7.3.68)$$

and

$$P_{\rm out}^{\rm max}(2\omega) = (1/2S)[(2g_o L')^{1/2} - (\gamma_L + \gamma_{NL})^{1/2}]^2. \qquad (1.7.3.69)$$

(1.7.3.69) shows that for the case where $\gamma_{\rm NL} \ll \gamma_L$ ($\gamma \simeq \gamma_L$), the maximum SH power is identically equal to the maximum fundamental power, (1.7.3.64), available from the same laser for the same value of loss, which, according to the previous definition of the intracavity efficiency, corresponds to an SHG conversion efficiency of 100%. $P_{\rm out}^{\rm max}(2\omega)$ strongly decreases as the losses $(\gamma_L + \gamma_{\rm NL})$ increase . Thus an efficient intracavity device requires the reduction of all losses at ω and 2ω to an absolute minimum.

(1.7.3.68) indicates that $K_{\rm opt}$ is independent of the operating power level of the laser, in contrast to the optimum transmitting mirror where $T_{\rm opt}$, given by (1.7.3.63), depends on the laser gain. $K_{\rm opt}$ depends only on the total losses and saturation parameter. For given losses, the knowledge of $K_{\rm opt}$ allows us to define the optimal parameters of the nonlinear crystal, in particular the figure of merit, $d^2_{\rm eff}/n_3^{2\omega}n_1^{\omega}n_2^{\omega}$ and the ratio $(L/w_o)^2$, in which the walk-off effect and the damage threshold must also be taken into account.

Some examples: a power of 1.1 W at 0.532 µm was generated in a $\rm TEM_{\it oo}$ c.w. SHG intracavity device using a 3.4 mm $Ba_2NaNb_5O_{15}$ crystal within a 1.064 µm Nd:YAG laser cavity (Geusic *et al.*, 1968). A power of 9.0 W has been generated at 0.532 µm using a more complicated geometry based on an Nd:YAG intracavity-lens folded-arm cavity configuration using KTP (Perkins & Fahlen, 1987). High-average-power SHG has also been demonstrated with output powers greater than 100 W at 0.532 µm in a KTP crystal inside the cavity of a diode side-pumped Nd:YAG laser (LeGarrec *et al.*, 1996).

For type-II phase matching, a rotated quarter waveplate is useful in order to reinstate the initial polarization of the fundamental waves after a round trip through the nonlinear crystal, the retardation plate and the mirror (Perkins & Driscoll, 1987).

If the nonlinear crystal surface on the laser medium side has a 100% reflecting coating at 2ω and if the other surface is 100% transmitting at 2ω, it is possible to extract the full SH power in one direction (Smith, 1970). Furthermore, such geometry allows us to avoid losses of the backward SH beam in the laser medium and in other optical components behind.

External-cavity SHG also leads to good results. The resonated wave may be the fundamental or the harmonic one. The corresponding theoretical background is detailed in Ashkin *et al.* (1966). For example, a bow-tie configuration allowed the generation of 6.5 W of $\rm TEM_{\it oo}$ c.w. 0.532 µm radiation in a 6 mm LiB_3O_5 (LBO) crystal; the Nd:YAG laser was an 18 W c.w. laser with an injection-locked single frequency (Yang *et al.*, 1991).

1.7.3.3.3. *Third harmonic generation (THG)*

Fig. 1.7.3.16 shows the three possible ways of achieving THG: a cascading interaction involving two $\chi^{(2)}$ processes, *i.e.* $\omega + \omega = 2\omega$ and $\omega + 2\omega = 3\omega$, in two crystals or in the same crystal, and direct THG, which involves $\chi^{(3)}$, *i.e.* $\omega + \omega + \omega = 3\omega$.

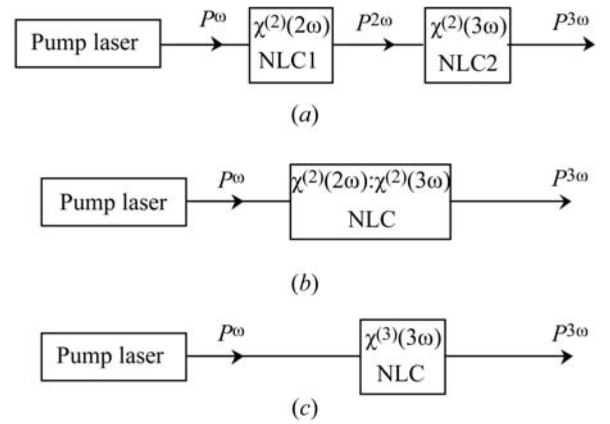

Fig. 1.7.3.16. Configurations for third harmonic generation. (*a*) Cascading process SHG ($\omega + \omega = 2\omega$): SFG ($\omega + 2\omega = 3\omega$) in two crystals NLC1 and NLC2 and (*b*) in a single nonlinear crystal NLC; (*c*) direct process THG ($\omega + \omega + \omega = 3\omega$) in a single nonlinear crystal NLC.

1.7.3.3.3.1. *SHG ($\omega + \omega = 2\omega$) and SFG ($\omega + 2\omega = 3\omega$) in different crystals*

We consider the case of the situation in which the SHG is phase-matched with or without pump depletion and in which the sum-frequency generation (SFG) process ($\omega + 2\omega = 3\omega$), phase-matched or not, is without pump depletion at ω and 2ω. All the waves are assumed to have a flat distribution given by (1.7.3.36) and the walk-off angles are nil, in order to simplify the calculations.

This configuration is the most frequently occurring case because it is unusual to get simultaneous phase matching of the two processes in a single crystal. The integration of equations (1.7.3.22) over Z for the SFG in the undepleted pump approximation with $E_1^{\omega}(Z_{\rm SFG} = 0) = E_1^{\omega}(L_{\rm SHG})$, $E_2^{2\omega}(Z_{\rm SFG} = 0) = E_2^{2\omega}(L_{\rm SHG})$ and $E_3^{3\omega}(Z_{\rm SFG} = 0) = 0$, followed by the integration over the cross section leads to

$$P^{3\omega}(L_{\rm SFG}) = B_{\rm SFG}[aP^{\omega}(L_{\rm SHG})]P^{2\omega}(L_{\rm SHG})\frac{L^2_{\rm SFG}}{w_o^2}\sin c^2\frac{\Delta k_{\rm SFG}L_{\rm SFG}}{2} \quad ({\rm W})$$

with

$$B_{\rm SFG} = \frac{72\pi}{\varepsilon_o c}\frac{2N-1}{N}\frac{d^2_{\rm eff}}{\lambda^2_{\omega}}\frac{T_3^{3\omega}T_1^{\omega}T_2^{2\omega}}{n_3^{3\omega}n_1^{\omega}n_2^{2\omega}} \quad ({\rm W}^{-1})$$

$$a = 1 \text{ for type-I SHG}, \quad a = \tfrac{1}{2} \text{ for type-II SHG}. \qquad (1.7.3.70)$$

$P^{\omega}(L_{\rm SHG})$ and $P^{2\omega}(L_{\rm SHG})$ are the fundamental and harmonic powers, respectively, at the exit of the first crystal. $L_{\rm SHG}$ and $L_{\rm SFG}$ are the lengths of the first and the second crystal, respectively. $\Delta k_{\rm SFG} = k^{3\omega} - (k^{\omega} + k^{2\omega})$ is the SFG phase mismatch. λ_{ω} is the fundamental wavelength. The units and other parameters are as defined in (1.7.3.42).

For type-II SHG, the fundamental waves are polarized in two orthogonal vibration planes, so only half of the fundamental power can be used for type-I, -II or -III SFG ($a = 1/2$), in contrast to type-I SHG ($a = 1$). In the latter case, and for type-I SFG, it is necessary to set the fundamental and second harmonic polarizations parallel.

The cascading conversion efficiency is calculated according to (1.7.3.61) and (1.7.3.70); the case of type-I SHG gives, for example,

$$\begin{aligned}\eta_{\rm THG}(L_{\rm SHG}, L_{\rm SFG}) &= \frac{P^{3\omega}(L_{\rm SFG})}{P^{\omega}_{\rm tot}(0)}\\ &= B_{\rm SFG}(T^{\omega})^4P^{\omega}_{\rm tot}(0)\tanh^2(\Gamma L_{\rm SHG})\\ &\quad\times {\rm sech}^2(\Gamma L_{\rm SHG})\frac{L^2_{\rm SFG}}{w_o^2}\sin c^2\left(\frac{\Delta k_{\rm SFG}L_{\rm SFG}}{2}\right),\end{aligned} \qquad (1.7.3.71)$$

where Γ is as in (1.7.3.59).

(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; ${\rm sech}(m)\to 1$, $\tanh(m)\to m$, and so we have

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

with

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

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, $\Delta k_{\rm SFG}\neq 0$,

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

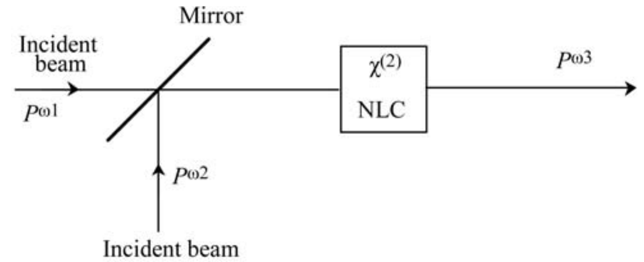

Fig. 1.7.3.17. Frequency up-conversion process $\omega_1 + \omega_2 = \omega_3$. The beam at ω_1 is mixed with the beam at ω_2 in the nonlinear crystal NLC in order to generate a beam at ω_3. $P^{\omega_1,\omega_2,\omega_3}$ are the different powers.

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

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

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

references