International Tables for Crystallography (2013). Vol. D, Section 1.7.3.3.2.4, pp. 208–209.

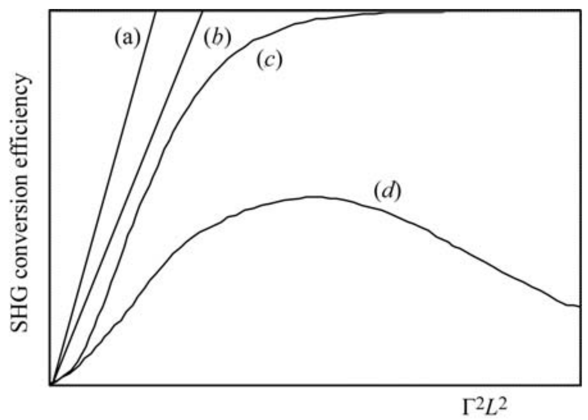

Fig. 1.7.3.15. Schematic SHG conversion efficiency for different situations of pump depletion and dephasing. (*a*) No depletion, no dephasing, $\eta = \Gamma^2 L^2$; (*b*) no depletion with constant dephasing δ, $\eta = \Gamma^2 L^2 \sin c^2 \delta$; (*c*) depletion without dephasing, $\eta = \tanh^2(\Gamma L)$; (*d*) depletion and dephasing, $\eta = \eta_m \mathrm{sn}^2(\Gamma L/v_b, v_b^4)$.

intensities of the two fundamental beams $I_1^\omega(X, Y, 0)$ and $I_2^\omega(X, Y, 0)$, which are not necessarily equal, are taken into account (Eimerl, 1987): the $\tanh^2$ function is valid only if perfect phase matching is achieved and if $I_1^\omega(X, Y, 0) = I_2^\omega(X, Y, 0)$, these conditions being never satisfied in real cases.

The situations described above are summarized in Fig. 1.7.3.15.

We give the example of type-II SHG experiments performed with a 10 Hz injection-seeded single-longitudinal-mode ($N = 1$) 1064 nm Nd:YAG (Spectra-Physics DCR-2A-10) laser equipped with super Gaussian mirrors; the pulse is 10 ns in duration and is near a Gaussian single-transverse mode, the beam radius is 4 mm, non-focused and polarized at $\pi/4$ to the principal axes of a 10 mm long KTP crystal ($L\delta\theta$ = 15 mrad cm, $L\delta\varphi$ = 100 mrad cm). The fundamental energy increases from 78 mJ (62 MW cm^{-2}) to 590 mJ (470 MW cm^{-2}), which correponds to the damage of the exit surface of the crystal; for each experiment, the crystal was rotated in order to obtain the maximum conversion efficiency. The peak power SHG conversion efficiency is estimated from the measured energy conversion efficiency multiplied by the ratio between the fundamental and harmonic pulse duration ($\tau_\omega/\tau_{2\omega} = 2^{1/2}$). It increases from 50% at 63 MW cm^{-2} to a maximum value of 85% at 200 MW cm^{-2} and decreases for higher intensities, reaching 50% at 470 MW cm^{-2} (Boulanger, Fejer *et al.*, 1994).

The integration of the intensity profiles (1.7.3.58) and (1.7.3.60) is obvious in the case of incident fundamental beams with a flat energy distribution (1.7.3.36). In this case, the fundamental and harmonic beams inside the crystal have the same profile and radius as the incident beam. Thus the powers are obtained from formulae (1.7.3.58) and (1.7.3.60) by expressing the intensity and electric field modulus as a function of the power, which is given by (1.7.3.38) with $m = 1$.

For a Gaussian incident fundamental beam, (1.7.3.37), the fundamental and harmonic beams are not Gaussian (Eckardt & Reintjes, 1984; Pliszka & Banerjee, 1993).

All the previous intensities are the peak values in the case of pulsed beams. The relation between average and peak powers, and then SHG efficiencies, is much more complicated than the ratio $\tau^{2\omega}/\tau^\omega$ of the undepleted case.

1.7.3.3.2.4. *Resonant SHG*

When the single-pass conversion efficiency SHG is too low, with c.w. lasers for example, it is possible to put the nonlinear crystal in a Fabry–Perot cavity external to the pump laser or directly inside the pump laser cavity, as shown in Figs. 1.7.3.6(*b*) and (*c*). The second solution, described later, is generally used because the available internal pump intensity is much larger.

We first recall some basic and simplified results of laser cavity theory without a nonlinear medium. We consider a laser in which one mirror is 100% reflecting and the second has a transmission T at the laser pulsation ω. The power within the cavity, $P_{\rm in}(\omega)$, is evaluated at the steady state by setting the round-trip saturated gain of the laser equal to the sum of all the losses. The output laser cavity, $P_{\rm out}(\omega)$, is given by (Siegman, 1986)

$$P_{\rm out}(\omega) = TP_{\rm in}(\omega)$$

with

$$P_{\rm in}(\omega) = \frac{2g_o L' - (\gamma + T)}{2S(T + \gamma)}. \qquad (1.7.3.62)$$

L' is the laser medium length, $g_o = \sigma N_o$ is the small-signal gain coefficient per unit length of laser medium, σ is the stimulated-emission cross section, N_o is the population inversion without oscillation, S is a saturation parameter characteristic of the nonlinearity of the laser transition, and $\gamma = \gamma_L = 2\alpha_L L' + \beta$ is the loss coefficient where α_L is the laser material absorption coefficient per unit length and β is another loss coefficient including absorption in the mirrors and scattering in both the laser medium and mirrors. For given g_o, S, α_L, β and L', the output power reaches a maximum value for an optimal transmission coefficient $T_{\rm opt}$ defined by $[\partial P_{\rm out}(\omega)/\partial T]_{T_{\rm opt}} = 0$, which gives

$$T_{\rm opt} = (2g_o L'\gamma)^{1/2} - \gamma. \qquad (1.7.3.63)$$

The maximum output power is then given by

$$P_{\rm out}^{\max}(\omega) = (1/2S)[(2g_o L')^{1/2} - \gamma^{1/2}]^2. \qquad (1.7.3.64)$$

In an intracavity SHG device, the two cavity mirrors are 100% reflecting at ω but one mirror is perfectly transmitting at 2ω. The presence of the nonlinear medium inside the cavity then leads to losses at ω equal to the round-trip-generated second harmonic (SH) power: half of the SH produced flows in the forward direction and half in the backward direction. Hence the highly transmitting mirror at 2ω is equivalent to a nonlinear transmission coefficient at ω which is equal to twice the single-pass SHG conversion efficiency $\eta_{\rm SHG}$.

The fundamental power inside the cavity $P_{\rm in}(\omega)$ is given at the steady state by setting, for a round trip, the saturated gain equal to the sum of the linear and nonlinear losses. $P_{\rm in}(\omega)$ is then given by (1.7.3.62), where T and γ are (Geusic *et al.*, 1968; Smith, 1970)

$$T = 2\eta_{\rm SHG} = [P_{\rm out}(2\omega)/P_{\rm in}(\omega)] \qquad (1.7.3.65)$$

and

$$\gamma = \gamma_L + \gamma_{NL}. \qquad (1.7.3.66)$$

$\eta_{\rm SHG}$ is the single-pass conversion efficiency. γ_L and γ_{NL} are the loss coefficients at ω of the laser medium and of the nonlinear crystal, respectively. L is the nonlinear medium length. The two faces of the nonlinear crystal are assumed to be antireflection-coated at ω.

In the undepleted pump approximation, the backward and forward power generated outside the nonlinear crystal at 2ω is

$$P_{\rm out}(2\omega) = 2KP_{\rm in}^2(\omega) \qquad (1.7.3.67)$$

with

$$K = B(L^2/w_o^2)\sin c^2(\Delta kL/2),$$

where

$$B = \frac{32\pi}{\varepsilon_o c}\frac{2N-1}{N}\frac{d_{\rm eff}^2}{\lambda_\omega^2}\frac{T_3^{2\omega}T_1^\omega T_2^\omega}{n_3^{2\omega}n_1^\omega n_2^\omega} \quad ({\rm W}^{-1}).$$

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

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

(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%. $P^{\rm max}_{\rm out}(2\omega)$ 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, $d^2_{\rm eff}/n^{2\omega}_3 n^{\omega}_1 n^{\omega}_2$ 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 $\rm TEM_{oo}$ c.w. SHG intracavity device using a 3.4 mm $\rm Ba_2NaNb_5O_{15}$ 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 $\rm TEM_{oo}$ c.w. 0.532 µm radiation in a 6 mm $\rm LiB_3O_5$ (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.* $\omega + \omega + \omega = 3\omega$.

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-

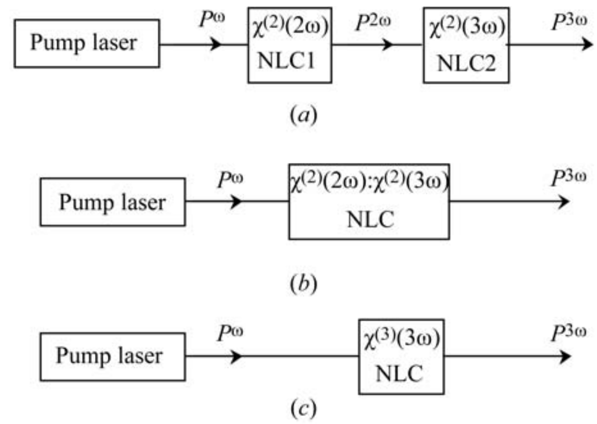

Fig. 1.7.3.16. Configurations for third harmonic generation. (*a*) Cascading process SHG $(\omega + \omega = 2\omega)$: SFG $(\omega + 2\omega = 3\omega)$ in two crystals NLC1 and NLC2 and (*b*) in a single nonlinear crystal NLC; (*c*) direct process THG $(\omega + \omega + \omega = 3\omega)$ in a single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

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 $E^{\omega}_1(Z_{\rm SFG} = 0) = E^{\omega}_1(L_{\rm SHG})$, $E^{2\omega}_2(Z_{\rm SFG} = 0) = E^{2\omega}_2(L_{\rm SHG})$ and $E^{3\omega}_3(Z_{\rm SFG} = 0) = 0$, followed by the integration over the cross section leads to

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

with

$$B_{\rm SFG} = \frac{72\pi}{\varepsilon_o c} \frac{2N-1}{N} \frac{d^2_{\rm eff}}{\lambda^2_{\omega}} \frac{T^{3\omega}_3 T^{\omega}_1 T^{2\omega}_2}{n^{3\omega}_3 n^{\omega}_1 n^{2\omega}_2} \quad ({\rm W}^{-1})$$
$$a = 1 \text{ for type-I SHG}, \quad a = \tfrac{1}{2} \text{ for type-II SHG}. \qquad (1.7.3.70)$$

$P^{\omega}(L_{\rm SHG})$ 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,

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

where Γ is as in (1.7.3.59).