

Fig. 4.1. Four Wave Mixing schemes. (a) The interaction between three waves at ω_1 , ω_2 and ω_3 generate a fourth wave at $\omega_4 = \omega_1 + \omega_2 + \omega_3$. (b) One single intense pump beam at ω_4 can generate 3 waves at ω_1 , ω_2 and ω_3 .

4.3 Four-Wave Mixing

Third order nonlinear interactions involve interaction between 4 waves at ω_1 , ω_2 , ω_3 and $\omega_4 = \omega_1 + \omega_2 + \omega_3$. Hereafter, we study different Four-Wave Mixing (FWM) configurations, non-degenerate and degenerate in frequencies, through lossless nonlinear materials with a purely real $\chi^{(3)}$ nonlinear susceptibility (and a purely real linear susceptibility). As a consequence, interactions will not be accompanied by any transfer of energy between the interacted waves and the nonlinear material. As for the 2nd order nonlinear effects, one could easily show that FWM interactions in lossless material follow the energy conservation relation:

$$\hbar\omega_1 + \hbar\omega_2 + \hbar\omega_3 = \hbar\omega_4.$$

Similarly, the derivation of nonlinear wave equations would give the following phase matching condition:

$$\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{k}_3 = \mathbf{k}_4,$$

which is equivalent to a law of momentum conservation.

4.3.1 Generation of UV and IR beams

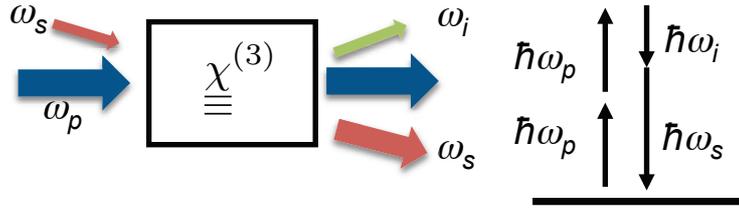
Figure 4.1 schematically illustrates (a) the capability for three incident waves at ω_1 , ω_2 and ω_3 to generate a fourth wave at $\omega_4 = \omega_1 + \omega_2 + \omega_3$, and (b) for one single intense pump beam at ω_4 to generate 3 waves at ω_1 , ω_2 and ω_3 . These interactions are governed by the phase matching condition $\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{k}_3 = \mathbf{k}_4$, which determines the directions of propagation for the 4 waves (in relation with the dispersion property of the nonlinear material). Whereas the interaction described in Fig. 4.1(a), which could serve to generate a UV beam from IR pump beams, can be theoretically described solving nonlinear wave equations, the Fig. 4.1(b) interaction requires a quantum description. Following the quantization of the optical electromagnetic waves, one could show that, through the interaction, one photon at ω_4 is annihilated to generate simultaneously 3 photons respectively at ω_1 , ω_2 and ω_3 .

These third order interactions find some interests in centro-symmetric material since their $\chi^{(2)}$ vanishes. Now, one has to underline the inherently weak efficiency of these nonlinear effects, since $\chi^{(3)}EEE \ll \chi^{(2)}EE$ for nonresonant interactions. A way to enhance the $\chi^{(3)}$ nonlinear susceptibility consists in using a quasi-resonant interaction as depicted in Figure 4.1(a) for which $\hbar\omega_1 + \hbar\omega_2 \simeq \hbar\omega_{ab}$, the energy transition between energy states $|a\rangle$ and $|b\rangle$.

4.3.2 Optical parametric amplification through FWM in an optical fiber

In the following, we illustrate the optical amplification of a signal ω_s using either two pump beams ω_{p1} and ω_{p2} , or one pump beam ω_p , through the interaction depicted in Figure 4.2 where

Fig. 4.2. 3rd Order optical parametric amplification : a signal beam ω_s is amplified through the interaction in a $\chi^{(3)}$ material with one single intense pump beam at ω_p . The interaction follows the energy conservation relation $\hbar\omega_p + \hbar\omega_p = \hbar\omega_s + \hbar\omega_i$, with ω_i the frequency of the idler beam that accompanies the amplification of the signal beam.



the beam frequencies follow the relation $\hbar\omega_p + \hbar\omega_p = \hbar\omega_s + \hbar\omega_i$. It is anticipated that the amplification of the signal beam ω_s is accompanied by the generation of a third beam at ω_i , referred to as the idler beam.

Optical fiber parametric amplifier: We next take the example of a parametric amplifier realized in a length of optical fiber. Typically the fiber is made in silica. Although the value of $\chi^{(3)}$ in silica remains rather small, significant amplification can be achieved in practice by using a long interaction length easily achievable in optical fibers. In addition, it would give the opportunity to play with nonlinear wave equations that has been derived in case of waveguides (see section 2.5.6).

The complex amplitude related to the three waves is written:

$$\mathbf{E}_m(\mathbf{r}, z) = \mathbf{e}_m \phi_m^q(\mathbf{r}) A_m(z) e^{i\beta_q(\omega_m)z}, \quad (4.2)$$

with $m = p, s$ or i . The three waves propagate inside a waveguide along the z direction, along which the waveguide is invariant. The mode field distribution related to each wave is characterized by the wavevector $\beta_p(\omega_m)$, the polarization state \mathbf{e}_m and the spatial distribution $\phi_m^p(\mathbf{r})$. Assuming that the set of transverse modes form a complete base of normalized and orthogonal modes, we have the relation (orthogonality between the transverse modes):

$$\int \int \phi_m^q(\mathbf{r}) \phi_m^{q'}(\mathbf{r})^* d^2\mathbf{r} = \delta_{qq'}.$$

Using the nonlinear wave equation (2.59), one can write three coupled wave equation at ω_p , ω_s and ω_i :

$$\begin{aligned} \frac{dA_p}{dz} &= \frac{i\omega_p}{2n_p c} 3\xi_{p-pp-p} \chi_{eff}^{(3)} |A_p|^2 A_p \\ \frac{dA_s}{dz} &= \frac{i\omega_s}{2n_s c} 3\chi_{eff}^{(3)} \left[2\xi_{p-ps-s} |A_p|^2 A_s + \xi_{ppi-s} A_p^2 A_i^* e^{i\Delta\beta z} \right] \\ \frac{dA_i}{dz} &= \frac{i\omega_i}{2n_i c} 3\chi_{eff}^{(3)} \left[2\xi_{p-pi-i} |A_p|^2 A_i + \xi_{p-ps-i} A_p^2 A_s^* e^{i\Delta\beta z} \right] \end{aligned} \quad (4.3)$$

As underlined by the right hand side term of wave equation (2.59), the modification of the envelope field is governed by the spatial overlapping between the spatial dependent nonlinear polarization term $\mathbf{P}^{(NL)}(\mathbf{r}, \omega)$ and the spatial distribution of the mode $\phi_m^p(\mathbf{r})$. As a consequence, the three wave equations are expressed with overlapping coefficients, which are given by:

$$\xi_{ijk-l} = \frac{\int \phi_i \phi_j \phi_k \phi_l^* d^2\mathbf{r}}{\int \phi_l \phi_l^* d^2\mathbf{r}} \quad \text{and} \quad \xi_{i-jk-l} = \frac{\int \phi_i \phi_j^* \phi_k \phi_l^* d^2\mathbf{r}}{\int \phi_l \phi_l^* d^2\mathbf{r}}, \quad (4.4)$$

where the indices i, j, k , and l refer to any of the interacted waves indices, p, s or i . Subsequently, we consider the propagation in a single mode fiber. The variation in the confinement for the

three waves is neglected and the overlapping coefficients are taken equal. Finally, the interaction efficiency is governed by the phase matching term:

$$\Delta\beta = 2\beta_p - \beta_s - \beta_i. \quad (4.5)$$

Note that nonlinear wave equations (4.3) assume that signal and idler wave intensities are much weaker than the pump intensity.

We start by deriving the wave equation for the pump:

$$\frac{dA_p}{dz} = i\gamma P_p A_p,$$

which has been expressed in terms of the pump power $P_p = 2nc\epsilon_0|A_p|^2 \int \int \phi_i \phi_i^* d^2\mathbf{r}$ and the parametric gain coefficient γ :

$$\gamma = \frac{3\omega_p}{4\epsilon_0 n_p^2 c} \frac{\int |\phi_p|^4 d^2\mathbf{r}}{(\int |\phi_p|^2 d^2\mathbf{r})^2} \chi_{eff}^{(3)} \quad (4.6)$$

Under a parametric regime, i.e. neglecting the pump depletion, the pump wave evolution follows

$$\begin{aligned} A_p(z) &= A_p(0) e^{i\gamma P_p z} \\ &= |A_p(0)| e^{i\theta} e^{i\gamma P_p z}, \end{aligned}$$

where θ is the linear phase related to the pump wave $A_p(z=0)$. The solution shows that, under parametric interaction, the undepleted pump wave experiences a nonlinear phase shift $\Phi_{NL}(z) = \gamma P_p z$. This nonlinear effect refers to *Optical Kerr Effect* that is studied in more details in section 4.4.

Substituting the pump wave evolution in wave equations (4.3), one can re-write wave equations for signal idler waves:

$$\frac{dA_s}{dz} = i\gamma P_p \frac{n_p \omega_s}{\omega_p n_s} \left[2A_s + A_i^* e^{2i\theta} e^{i(\Delta\beta + 2\gamma P_p)z} \right] \quad (4.7)$$

$$\frac{dA_i^*}{dz} = -i\gamma P_p \frac{n_p \omega_i}{\omega_p n_i} \left[2A_i^* + A_s e^{-2i\theta} e^{-i(\Delta\beta + 2\gamma P_p)z} \right]. \quad (4.8)$$

In order to simplify those coupled equations one can introduce new variables:

$$B_s(z) = A_s(z) \exp\left(-2i\gamma P_p \frac{n_p \omega_s}{\omega_p n_s}\right) \quad \text{and} \quad B_i^*(z) = A_i^*(z) \exp\left(+2i\gamma P_p \frac{n_p \omega_i}{\omega_p n_i}\right),$$

leading to the set of coupled equations:

$$\frac{dB_s}{dz} = i\gamma P_p \frac{n_p \omega_s}{\omega_p n_s} e^{2i\theta} B_i^*(z) e^{+iK'z} \quad (4.9)$$

$$\frac{dB_i^*}{dz} = -i\gamma P_p \frac{n_p \omega_i}{\omega_p n_i} e^{-2i\theta} B_s(z) e^{-iK'z}. \quad (4.10)$$

with $K' = \Delta\beta + 2\gamma P_p \left[1 - \frac{n_p}{\omega_p} \left(\frac{\omega_s}{n_s} + \frac{\omega_i}{n_i}\right)\right]$. Actually, the previous coupled equations take a form similar to those derived for the parametric amplification in a $\chi^{(2)}$ material (section 3.5), helping in deriving the solutions in case of parametric amplification in a $\chi^{(3)}$ material.

Assuming the boundary conditions $A_i(z = 0) = 0$ and $A_s(z = 0) = A_{s0}$, the solutions for signal and idler optical power evolutions are:

$$P_s(z) = P_s(0) \left[1 + \frac{\Gamma'^2}{g'^2} \sinh^2(g'z) \right] \quad (4.11)$$

$$P_i(z) = \frac{\omega_i n_s}{n_i \omega_s} P_s(0) \frac{\Gamma'^2}{g'^2} \sinh^2(g'z), \quad (4.12)$$

with $\Gamma' = \gamma P_p \frac{n_p}{\omega_p} \sqrt{\frac{\omega_i \omega_s}{n_i n_s}}$ and $g'^2 = \Gamma'^2 - \frac{K'^2}{4}$, the parametric gain.

The amplification factor for the signal can be directly derived from (4.11):

$$G(z) = \frac{\Gamma'^2}{g'^2} \sinh^2(g'z). \quad (4.13)$$

As a conclusion, an incident signal wave at ω_s is subject to amplification through its interaction with a pump beam at ω_p in a $\chi^{(3)}$ nonlinear material. Assuming a lossless material, this amplification is accompanied by the generation of an idler beam at $\omega_i = 2\omega_p - \omega_s$. The amplification factor is directly proportional to the pump intensity² and depends on the phase matching condition given by $K' = \Delta\beta + 2\gamma P_p \left[1 - \frac{n_p}{\omega_p} \left(\frac{\omega_s}{n_s} + \frac{\omega_i}{n_i} \right) \right]$. As it will be underlined below, this phase matching condition is influenced by the Optical Kerr Effect, through the term proportional to γP_p . Considering that $\frac{n_p}{\omega_p} \simeq \frac{n_s}{\omega_s} \simeq \frac{n_i}{\omega_i}$, the phase matching condition can be simplified in:

$$\begin{aligned} K' &= \Delta\beta + 2\gamma P_p \left[1 - \frac{n_p}{\omega_p} \left(\frac{\omega_s}{n_s} + \frac{\omega_i}{n_i} \right) \right] \\ &\simeq \Delta\beta - 2\gamma P_p. \end{aligned} \quad (4.14)$$

In the case of a perfect phase matching condition $K' = 0$, the amplification factor reaches its maximum value for $K' = 0$:

$$\begin{aligned} G_{max}(z) &\simeq \sinh^2(\gamma P_p z) \\ &\simeq \sinh^2(\Phi_{NL}(z)) \end{aligned} \quad (4.15)$$

since $g' = \Gamma'$. We have assumed that $\frac{n_p}{\omega_p} \simeq \frac{n_s}{\omega_s} \simeq \frac{n_i}{\omega_i}$.

Comments:

- The phase matching condition is described by a linear term, $\Delta\beta$ the phase mismatch between the interacted waves, and a nonlinear effect related to the nonlinear phase $\Phi_{NL}(z) = \gamma P_p z$ experienced by the pump wave. As it will be explained in the next section, the pump intensity modifies the refractive index of the material, which directly influenced the phase matching condition.
- Concerning the phase mismatch $\Delta\beta = 2\beta_p - \beta_s - \beta_i$ between the interacted waves, we show its relation with the dispersion coefficient β_2 of the material (or the waveguide). Indeed, one can write $\omega_s = \omega_p - \Omega$, meaning that $\omega_i = \omega_p + \Omega$. if we consider a narrow spectral interval Ω between the interacted waves, i.e. $\Omega \ll \omega_p$, the Taylor's expansions for β_s and β_i show that $\Delta\beta \simeq -\beta_2 \Omega^2$.

²The amplification factor depends on the quantity γP_p and, one could note that the expression (4.6) for γ depends on the mode field distribution of the waves.

Neglecting the Optical Kerr Effect, parametric amplification would require the propagation through a waveguide with a zero dispersion, $\beta_2 = 0$. However, the phase matching is fulfilled since $K' = 0$, implying $\Delta\beta = 2\gamma P_p(z = 0)$. Now, depending on the sign of the nonlinear coefficient $\chi_{eff}^{(3)}$ (equivalently of γ), phase matching can only be fulfilled with dispersion coefficient β_2 which takes an opposite sign. Considering for instance, parametric amplification in a silica fiber, for which $\gamma > 0$, amplification implies the propagation in anomalous dispersion regime with $\beta_2 < 0$ ³.

- Parametric amplification occurs only if $g'^2 > 0$, or $\Gamma'^2 > K'^2/4$, which implies the following condition for the linear phase mismatch: $0 < \Delta\beta < 4\gamma P_p$. The maximum value for amplification is reached for $\Delta\beta = 2\gamma P_p$, coinciding with $K'^2 = 0$.

4.3.3 Optical Parametric Fluorescence Effect

We next consider the FWM depicted in Figure 4.2, but where we suppose that only one pump beam is incident on the $\chi^{(3)}$ material. The solutions (4.11) for the FWM wave equations show that, with the boundary conditions $A_s(z = 0) = 0$ and $A_i(z = 0) = 0$, the signal and idler power remains null: $P_s(z) = 0$ and $P_i(z) = 0$. However, and similarly to the parametric fluorescence effect observed in a $\chi^{(2)}$ material, signal and idler photons are generated at frequencies which minimize the phase matching condition for the material (or the waveguide) used in the experiment. A complete description of such an effect requires the optical field quantization at ω_s and ω_i , with an approach similar to that of a $\chi^{(2)}$ fluorescence effect.

4.3.4 Frequency degenerated FWM : phase conjugate mirror

See Tutorial.

³ $\beta_2 < 0$ coincides with a positive dispersion coefficient D .