Module 4 : Third order nonlinear optical processes

Lecture 19 : General Theory of four-ware-mixing

Objectives

In this lecture we will illustrate the general features and formalism of the four-wave-mixing processes subdivided into four particular cases. Specific examples of each of these will be encountered in subsequent lectures.

General Theory of four-ware-mixing

In four-wave-mixing phenomena, three pump fields at frequencies ω_1, ω_2 and ω_3 interact with an optical medium characterized by third order nonlinear susceptibility and produce a polarization $P_{NL}^{(3)}(\omega_s)$ oscillating at their mixed frequency $\omega_s = \omega_1 \pm \omega_2 \pm \omega_3$. which then radiates the fourth field, the signal field at frequency ω_s. For the sake of simplicity and without much loss of the generality, we assume

1. Participating electromagnetic fields to be plane monochromatic.
2. Undepleted pump fields.
3. Signal field propagating along z-direction.
4. Nonlinear optical medium occupying the positive half space $z \geq 0$ to be cubic or isotropic.

We will be discussing the following four cases.

Case # 1.

We consider three monochromatic pump fields $E_1(r,t)$, $E_2(r,t)$ and $E_3(r,t)$, each one of the form

$$\vec{E}_q(\vec{r},t) = \hat{e}_q E_q(z) e^{i(\vec{k}_q\cdot\vec{r}-\omega_q t)} \tag{19.1}$$

where E_q is the field amplitude and $\hat{e}_q$ is its polarization direction unit vector interacting in a nonlinear medium in a geometry shown in figure 19.1 and generate a fourth field, the signal field of the form

$$\vec{E}_s(\vec{r},t) = \hat{e}_s E_s(z) e^{i(\vec{k}_s\cdot\vec{r}-\omega_s t)} \tag{19.2}$$

with $\omega_s = \omega_1 + \omega_2 + \omega_3$.

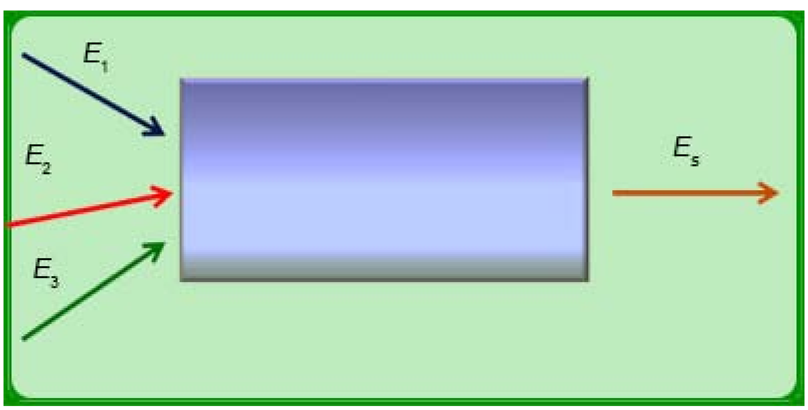

Figure 19.1

In slowly varying envelop approximation (SVEA) (see lecture 9 equation 9.19), the spatial evolution of the signal field is described by

$$\frac{dE_s}{dz} = -i\,\frac{\omega_s^2\mu_0}{2k_s}\left(\vec{P}^{(3)}(z,\omega_s)\cdot\hat{e}_s\right)e^{ik_s z}$$

$$= -\frac{i\omega_s^2\mu_0}{2k_s}\sum_i P_i^{(3)}(z,\omega_s)\,e_{si}e^{ik_s z} \tag{19.3}$$

Where we have used

$$\vec{P}^{(3)}(z,\omega_s)\cdot\hat{e}_s = \sum_{\Gamma} P^{(3)}(z,\omega_s)\,e_{si} \tag{19.4}$$

as the component of the nonlinear polarization which is effective for the generation of the signal.

One can express the nonlinear polarization in terms of pump fields as

$$\sum_i P_i^{(3)}(z,\omega_s)\,e_{si} = \epsilon_0 D\sum_{jkl}\chi_{ijkl}^{(3)}(-\omega_s;\omega_1,\omega_2,\omega_3)\,e_{si}e_{1j}e_{2k}e_{3l}E_1(z)E_2(z)E_3(z)\exp\left(-i\left(\vec{k}_1+\vec{k}_2+\vec{k}_3\right)\cdot\vec{r}\right)$$

$$= \epsilon_0\chi_{eff}^{(3)}E_1(z)E_2(z)E_3(z)\exp\left(-i\left(\vec{k}_1+\vec{k}_z+\vec{k}_3\right)\cdot\vec{r}\right) \tag{19.5}$$

D is she number of distinct permutations of the pump field frequencies. Here we have defined the effective susceptibility as

$$\chi_{eff}^{(3)} = D\sum_{jkl}\chi_{ijkl}^{(3)}(-\omega_s;\omega_1,\omega_2,\omega_3)\,e_{si}e_{1j}e_{2k}e_{3l} \tag{19.6}$$

Thus we can write the signal field evolution equation as

$$\frac{dE_s}{dz} = -\frac{i\omega_s^2\mu_0\epsilon_0}{2k_s}\chi_{eff}^{(3)}E_1(z)E_2(z)E_3(z)\,e^{-i\left(\Delta\vec{k}\cdot\hat{e}_z\right)z} \tag{19.7}$$

$\Delta\vec{k} = \vec{k}_s - \vec{k}_1 - \vec{k}_2 - \vec{k}_3$ is the wave vector or phase mismatch.

using the initial condition $E_s(z=0)=0$, the signal field can be obtained upon integration of the above differential equation.

$$E_s(z) = \frac{\omega_s^2\mu_0\epsilon_0}{2k_s\left(\overrightarrow{\Delta k}\cdot\hat{e}_z\right)}\chi_{eff}^{(3)}E_1(0)E_2(0)E_3(0)\left[\exp\left(-i\left(\overrightarrow{\Delta k}\cdot\hat{e}_z\right)z\right)-1\right] \tag{19.8}$$

Since

$$\left[\exp\left(-i\left(\overrightarrow{\Delta k}\cdot\hat{e}_z\right)z\right)-1\right] = \frac{\sin\left(\frac{\left(\Delta\vec{k}\cdot\hat{e}_z\right)z}{2}\right)}{\left[\frac{\left(\Delta\vec{k}\cdot\hat{e}_z\right)z}{2}\right]} \tag{19.9}$$

equation 19.8 implies that the signal goes through its maximum and minimum intensity i.e.

(19.10)

$$\left|E_s(z)\right|^2 \propto \frac{\sin^2\left(\dfrac{\left(\Delta\vec{k}\cdot\hat{e}_z\right)z}{2}\right)}{\left[\dfrac{\left(\Delta\vec{k}\cdot\hat{e}_z\right)z}{2}\right]^2}$$

unless the phase matching condition $\Delta k = 0$ is satisfied.
In phase matched generation the signal intensity

$$\left|E_s(z)\right|^2 \sim z^2 \qquad (19.11)$$

In four-wave-mixing (FWM), it can be achieved by appropriate alignment of pump

Case # 2

We next consider the interaction geometry shown in figure 19.2 where the signal field E_s is degenerate with the pump field in its frequency $\omega_s = \omega_3$. That is to say that the two fields E_s and E_3 are in the same mode. We will assume that the other two pump fields $\vec{E}_1$, and $\vec{E}_2$ remain constant. In this case, one can readily write the signal field upon integration of the equation (19.7)

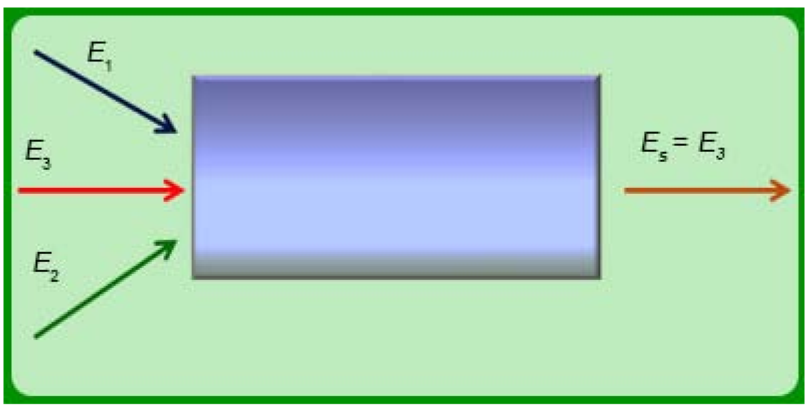

Figure 19.2

$$E_s(z) = E_s(0)\exp(g_s z) \qquad (19.12)$$

where

$$g_s(z) = \frac{\omega_s^2\mu_0\varepsilon_0}{2k_s\left(\vec{\Delta}k\cdot\hat{e}_z\right)}\chi_{eff}^{(3)}E_1(0)E_2(0)\left[\exp\left(-i\left(\Delta\vec{k}\cdot\hat{e}_z\right)z\right)-1\right] \qquad (19.13)$$

The real part of $g_s(z)$ stands for the gain/loss of the signal field propagating through the nonlinear medium.
For small value of *z*

$$E_2(0) = E_1^*(0) \qquad (19.14)$$

We get

$$\mathrm{Re}[g_s(z)] = \frac{\omega_s^2\mu_0\varepsilon_0}{2k_s}\mathrm{Im}\,\chi_{eff}^{(3)}\left|E_1(0)\right|^2 z \qquad (19.15)$$

This implies that the signal field grows as we will find in the case of stimulated Raman scattering in the (Lecture 27).

Case #3

We consider the interaction geometry shown in figure 19.3 with two weak counter propagating waves $\vec{E}_s(z)$ and $\vec{E}_3(z)$ and two pump fields $\vec{E}_1(0)$, and $\vec{E}_2(0)$ which, the latter remain constant.

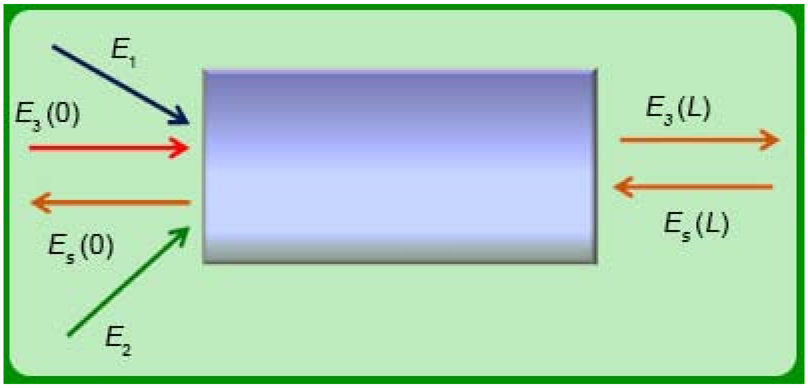

Figure 19.3

In this case, we will have to write one additional differential equation for the evolution of the field $\vec{E}_3(z)$. Remember that $\omega_3 = \omega_s - \omega_1 - \omega_2$

$$\frac{\mathrm{d}E_3}{\mathrm{d}z} = -\frac{\mathrm{i}\omega_s^2\mu_0\varepsilon_0}{2k_s}\chi_{eff}^{(3)}E_s(z)E_1^*(0)E_2^*(0)\mathrm{e}^{\mathrm{i}(\Delta\vec{k}\cdot\hat{e}_z)z} \tag{19.16}$$

Considering the phase matched case $\Delta k = 0$, the two equations can be decoupled and written as

$$\left(\frac{\mathrm{d}^2}{\mathrm{d}z^2} + \kappa^2\right)\begin{bmatrix} E_s \\ E_3 \end{bmatrix} = 0 \tag{19.17}$$

where

$$\kappa^2 = \frac{\omega_s^2\omega_3^2}{k_s k_3}\left|\frac{\chi_{eff}^{(3)}E_1E_2}{2c^2}\right|^2 \tag{19.18}$$

The situation here is similar to the backward parametric amplification where $\vec{E}_3(z)$ plays the role of idler.

With the initial conditions for $E_s(z=L)$ and $E_3(z=0)$ specified, we can obtain the following solutions for the signal and idler fields.

$$E_s(z=0) = \frac{E_s(L)}{\cos\kappa L} + \frac{\mathrm{i}A\omega_s}{|A|\omega_3}\sqrt{\frac{k_3}{k_s}}E_3(0)\tan\kappa L \tag{19.19}$$

$$E_3(z=L) = -\frac{\mathrm{i}}{A}\frac{|A|\omega_3}{\omega_s}\sqrt{\frac{k_s}{k_3}}E_s(L)\tan\kappa L + \frac{E_3(0)}{\cos\kappa L} \tag{19.20}$$

If $\kappa L \to \pi/2$ then $E_s(0)$ and $E_3(L)$ diverge which implies that even without one signal and idler initially being present, $E_s(0)$ and $E_3(L)$ can be generated from noise. Thus one realizes parametric oscillations. The necessary energy is supplied by the two pumps. In FWM such parametric oscillations are easily realizable. On the other hand in three-wave-mixing parametric oscillations in backward geometry are not possible due to the phase matching issues with only one pump.

Case # 4

Finally we consider the partially degenerate four-wave-mixing wherein all the four field wave vectors are parallel as shown in figure 19.4.

Figure 19.4

For the process $\omega_4 = \omega_3 + \omega_2 + \omega_1$ (either with $\omega_1 = \omega_2 = \omega_3$ or $\omega_1 = \omega_2 \neq \omega_3$) phase matching in this case is difficult. However in partially degenerate case, it is possible to achieve i.e. when $\omega_1 = \omega_2 = \omega_p$ is the pump wave frequency and

$$\omega_4 = 2\omega_p - \omega_3 \tag{19.21}$$

or

$$\omega_p = \frac{\omega_3 + \omega_4}{2} \tag{19.22}$$

In this case

$$\Delta k = k_4 + k_3 - k_2 - k_1 \tag{19.23}$$

$$= \frac{1}{c}\left[\omega_4 n_4 + \omega_3 n_3 - 2\omega_p n_p\right] \tag{19.24}$$

Such a propagation scenario occurs in optical fibers in the anomalous dispersion regime which leads to a modulation instability.

Let us assume that all waves propagate in $+z$ direction and $\omega_4 > \omega_3$.

An intense pump wave at

$$\omega_p = \omega_1 = \frac{\omega_3 + \omega_4}{2} \tag{19.25}$$

creates two symmetrical side bands at a ω_3 and ω_4 with frequency shifts of

$$\Omega_s = \omega_p - \omega_3 = \omega_4 - \omega_p \tag{19.26}$$

Down shifted ω_3 is called the Stokes and up shifted ω_4 the anti-Stokes frequency. If momentum conservation is fulfilled , ω_3 and ω_4 are spontaneously generated from the noise. It a weak signal is present initially, it will be amplified with simultaneous creation of the idler wave at ω_4.

Assuming that the pumps are undepleted, we can write down two differential equations for E_3 and E_4 as

$$\frac{dE_3}{dz} = -\frac{i\omega_3^2\mu_0\varepsilon_0}{2k_3}\chi_{eff}^{(3)}E_1^2(0)E_4^*(z)\,e^{-i\Delta kz} \tag{19.27}$$

$$\frac{dE_4}{dz} = -\frac{i\,\omega_4^2 \mu_0 \varepsilon_0}{2k_4} \chi_{eff}^{(3)} E_1^2(0) E_3^*(z)\, e^{i\Delta kz} \tag{19.28}$$

Elimination of say E_4 from equation (19.31) yields a second order differential equation

$$\frac{d^2 E_3}{dz^2} - i\Delta k \frac{dE_3}{dz} - K^2 E_3 = 0 \tag{19.29}$$

where we have defined.

$$K = \frac{\omega_3 \omega_4 \mu_0 \varepsilon_0}{2\left(\sqrt{k_3 k_4}\right)} \left|E_1(0)\right|^2 \tag{19.30}$$

Solution for this can readily written as

$$E_3(z) = \exp\left(i\left(\frac{\Delta kz}{2}\right)\right)\left(Ae^{g_+ z} + Be^{g_- z}\right) \tag{19.31}$$

Similarly, we can describe the solution for the other coupled wave as

$$E_4(z) = \exp\left(i\left(\frac{\Delta kz}{2}\right)\right)\left(Ce^{g_+ z} + De^{g_- z}\right) \tag{19.32}$$

with parametric gain defined by

$$g_\pm = \pm\sqrt{K^2 - \left(\frac{\Delta k}{2}\right)^2} \tag{19.33}$$

which suggests that each of the coupled waves splits in to two modes, one with gain g_+ and other with g_-. A similar case we will encounter in case of Stokes and anti-Stokes coupling in Raman scattering (Lecture 29).

Recap

In this lecture we have illustrated some general features and formalism of the four-wave-mixing processes subdivided into following four particular cases:

1. All the pump and signal fields with different frequency and propagation vectors.
2. Two pump fields with distinct frequency and propagation direction and other two fields in the same mode.
3. Two non-degenerate pump fields and the other two counter propagating fields of different frequencies. Possibility of Backward parametric amplification/oscillations is demonstrated here.
4. Case of partially degenerate four-wave-mixing.

Specific phenomena related to each of these cases will be discussed in subsequent lectures.