Module 2 : Nonlinear Frequency Mixing

Lecture 11 : Three Wave Mixing

Objectives

In this lecture we will look at

- Coupled wave equations for sum and difference frequency generation.
- Manley Rowe Relations.

When two different monochromatic coherent waves with frequencies $\omega_1 \text{ and } \omega_2$ are incident, the second order non-linear polarization has components at $2\omega_1, 2\omega_2, 0$ as before but now also at $\omega_1 \pm \omega_2$. These sum frequency generation $\left(SFG\!: \omega_1 + \omega_2 \rightarrow \omega_3 = \omega_1 + \omega_2\right)$ and difference frequency generation process $\left(DFG\!: \omega_3 - \omega_2 \rightarrow \omega_1 = \omega_3 - \omega_2\right)$ can all be described in terms of interaction between 3 waves. Let the 3 interacting frequencies be $\omega_p, \omega_s \text{ and } \omega_i.$ with $\omega_p = \omega_s + \omega_i.$ The three nonlinear polarizations are given by

$$P_\mu\left(\omega_p\right) = 2\,\varepsilon_0\,\chi^{(2)}_{\mu\alpha\beta}\left(-\omega_p, \omega_s, \omega_i\right) E_\alpha\left(\omega_s\right) E_\beta(\omega_i) \tag{11.1}$$

$$P_\alpha\left(\omega_s\right) = 2\,\varepsilon_0\,\chi^{(3)}_{\alpha\mu\beta}\left(-\omega_s, \omega_p, -\omega_i\right) E_\mu\left(\omega_p\right) E^*_\beta(\omega_i) \tag{11.2}$$

$$P_\beta\left(\omega_i\right) = 2\,\varepsilon_0\,\chi^{(3)}_{\beta\mu\alpha}\left(-\omega_i, \omega_p, -\omega_s\right) E_\mu\left(\omega_p\right) E^*_\alpha(\omega_s) \tag{11.3}$$

As for SHG, we need to write wave equations for $\vec{E}(\omega_p), \vec{E}(\omega_s) \; and \; \vec{E}(\omega_i)$ and then in the slowly varying envelope approximation (see e.g. eq 9.19) for the 3 complex amplitudes $A_p, A_s \; and \; A_i$. We write

$$\vec{E}(\omega_n) = \vec{a}_n\,\vec{A}_n\left(z\right)e^{ik_n z} \tag{11.4}$$

with the polarization vectors chosen to satisfy

$$k_n^{\,2}\,(\hat{z}\times\hat{z}\times\hat{a}_n) + \frac{\omega^2}{c^2}\overleftrightarrow{\varepsilon}(\omega_n).\,\hat{a}_n = 0 \text{ with } n = p, s \text{ or } i. \tag{11.5}$$

As in the earlier case we obtain an equation for the complex amplitudes $A_i, A_s \text{ and } A_p$. E.g.,

$$-2i\,k_p\,\frac{\partial A_p}{\partial z}\cos^2\alpha_p = \frac{{\omega_p}^2}{c^2}A_s A_i\,\overleftrightarrow{\chi}^{(2)}\left(-\omega_p, \omega_s, \omega_i\right)\vdots\,\hat{a}_p\hat{a}_s\hat{a}_i e^{-i\Delta kz} \tag{11.6}$$

with $\Delta k = k_p - k_s - k_i$. Then, using the overall permutation symmetry of the susceptibility tensor, one can write the three equations in terms of a single coupling coefficient. We obtain

$$\frac{\partial A_s^*}{\partial z} = -\frac{{\omega_1}^2 K}{k_s \cos^2\alpha_s}\;A_p^* A_i e^{-i\Delta kz} \tag{11.7}$$

$$\frac{\partial A_i^*}{\partial z} = -\frac{{\omega_i}^2 K}{k_i \cos^2\alpha_i}\;A_p^* A_s e^{-i\Delta kz} \tag{11.8}$$

$$\frac{\partial A_p}{\partial z} = -\frac{{\omega_p}^2 K}{k_p \cos^2\alpha_p}\;A_s A_i e^{-i\Delta kz} \tag{11.9}$$

with

$$K = 2\varepsilon_0\,\overleftrightarrow{\chi}^{(2)}\left(-\omega_3, \omega_1, \omega_2\right)\vdots\,\hat{a}_3\hat{a}_1\hat{a}_i \tag{11.10}$$

Then writing

$$A_i = \rho_i e^{i\phi_i} \qquad (11.11)$$

and separating the real and imaginary parts we obtain equations of motion for the 3 amplitudes and the 3 phases. For the amplitudes we get

$$\frac{\partial \rho_s}{\partial z} = -\frac{\omega_s{}^2 K}{k_s \cos^2 \alpha_s} \rho_p \rho_i \sin\theta \qquad (11.12)$$

and similar equations for ρ_i and ρ_p. For the phases we get

$$\frac{\partial \phi_s}{\partial z} = \frac{\omega_s{}^2 K}{k_s \cos^2 \alpha_s} \frac{\rho_i \rho_p}{\rho_s} \cos\theta \qquad (11.13)$$

$$\frac{\partial \phi_i}{\partial z} = \frac{\omega_i{}^2 K}{k_i \cos^2 \alpha_i} \frac{\rho_s \rho_p}{\rho_i} \cos\theta \qquad (11.14)$$

$$\frac{\partial \phi_p}{\partial z} = \frac{\omega_p{}^2 K}{k_p \cos^2 \alpha_p} \frac{\rho_1 \rho_2}{\rho_3} \cos\theta \qquad (11.15)$$

where

$$\theta = \Delta kz + \phi_p - \phi_s - \phi_i \qquad (11.16)$$

Here we have taken all three waves to propagate in the same direction z, polarization vectors are taken as real corresponding to linear polarization.

We should mention here that the phase change of an optical wave due to nonlinear coupling is an important manifestation of the intensity dependent refractive index or the Kerr nonlinearity which is a third order nonlinearity. Since this phase change also occurs in the coupled wave solutions it is sometimes seen as a cascaded second order nonlinearity in the sense that there is a change in phase of the fundamental beam due to coupling with the second harmonic which is created by conversion from the fundamental. Clearly this is not a new nonlinear mechanism and is fully included in the ABDP analysis. However, the consequences of this realization are important. To begin with, in media without inversion symmetry the two nonlinear mechanisms can interfere and provide a way of determining the sign of $\chi^{(3)}$. More important, this cascading nonlinearity can be much larger and give rise to strong effects such as spatial solitons and optical switching.

From eqs (11.7) to (11.9) it is easy to show that

$$\frac{\partial}{\partial z}\left(\left|A_s\right|^2 + \left|A_i\right|^2 + \left|A_i\right|^2\right) = 0 \qquad (11.17)$$

corresponding to the conservation of power in the frequency conversion process in a transparent medium. If we denote the total power per unit area as ***W***, we can define a scaled interaction length length

$$\zeta = z.\chi_{eff}^{(2)}\left(\frac{W\omega_p\omega_s\omega_i}{\varepsilon_0 c^3 n_p n_i n_s \cos\alpha_p \cos\alpha_s \cos\alpha_i}\right)^{1/2} \qquad (11.18)$$

Similarly we define the scaled amplitudes v_p, v_s and v_i with corresponding phases ϕ_p, ϕ_s **and** ϕ_i, respectively such that v_p^2, v_s^2 and v_i^2 are the fractional powers in the three waves.

It is important to emphasize that it is by virtue of overall permutation symmetry that the 3 processes namely generation of ω_p from ω_s and ω_i; or generation of ω_s from ω_p and ω_i are all described by the same susceptibility tensor. The evolution of the three amplitudes v_p, v_s and v_i is given by

(11.19)

$$\frac{dv_s}{d\zeta} = -\frac{\omega_s}{\omega_p} v_p v_i \sin\theta$$

$$\frac{dv_i}{d\zeta} = -\frac{\omega_i}{\omega_p} v_p v_s \sin\theta \tag{11.20}$$

$$\frac{dv_p}{d\zeta} = v_i v_s \sin\theta \tag{11.21}$$

and

$$\frac{d\theta}{d\zeta} = \Delta s + \cot\theta \frac{d}{d\zeta}\left(\ln u_p u_s u_i\right) \tag{11.22}$$

where θ is the relative phase

$$\theta = \Delta kz + \phi_p - \phi_i - \phi_s \tag{11.23}$$

and

$$\Delta s = \Delta k(z/\zeta) \tag{11.24}$$

is the scaled phase mismatch. v_s^2, v_i^2 and v_p^2 represents the fractional power in the signal, idler and pump waves, respectively with

$$v_i^2 + v_s^2 + v_p^2 = 1 \tag{11.25}$$

From eqs (62-64), one finds that

$$\frac{\omega_p}{\omega_s}\frac{d}{d\zeta}v_s^2 = \frac{\omega_p}{\omega_i}\frac{d}{d\zeta}v_i^2 = -\frac{d}{d\zeta}v_p^2 \tag{11.26}$$

implying that in nonlinear propagation

$$\delta v_s^2 / \omega_s = \delta v_i^2 / \omega_i = -\delta v_p^2 / \omega_p \tag{11.27}$$

These equations imply that the power in the signal and the idler waves grow or decrease together while that in pump beam decreases if signal wave is growing, and grows if the signal wave is decreasing. Eqs (11.27) are called the Manely – Rowe Relations. In terms of photon fluxes N_n in the three beams these imply that

$$\delta N_s = +\delta N_i = -\delta N_p \tag{11.28}$$

i.e. ***every photon lost in the pump wave implies a gain of one photon each in the signal and idler waves***.

The remaining equation for the pump wave amplitude is integrated the same way as for second harmonic case and will be discussed in the next lecture.

These solutions, of course, describe the full range of 3 wave mixing processes. Given initial amplitudes and phase of the 3 waves, we can determine them after a distance *z*. The direction of energy flow i.e. from or to the ω_p wave is determined by the relative phase θ, which becomes undefined if any wave has zero initial amplitude. If two of the three initial amplitudes are non zero, the third wave can only grow in amplitude and this determines the phase q.

In the small signal regime, this amplitude will grow as ~ z. The growth rate is also proportional to the two incident amplitudes and the effective non-linear coupling coefficient χ_{eff}^2. The evolution of the highest frequency wave (ω_p) is quite different from that of ω_i and ω_s. If initially wi and ws are present, ω_p can grow from zero. This is the sum frequency generation (SFG) processes. But unlike SHG the process does not continue in the same direction i.e. from ω_s, ω_i ω_p because for each photon of ω_p produced one is lost from ω_i and one from ws. In some finite distance, which depends on the photon fluxes in the two waves, one of the two input waves – the one with smaller photon flux- will be depleted fully. The growth of the pump wave has to stop then. For specificity let us take ω_S is the depleted wave. Now we have $I_S = 0$ and I_p and I_i are non zero. q is again undefined because ϕ_s is undefined. But $\frac{\partial u_s}{\partial \zeta}$ is non zero, so the signal wave will grow and Ip will reduce. This is now the difference frequency generation (DFG) since we are generating ω_S from ω_p and ω_i ($\omega_i = \omega_p - \omega_s$).

It is important to note that according to Manely Rowe relations as I_S increases, I_i also increases. This implies that as the signal beam grows from 0 initial amplitude, the idler wave is also amplified. The pump wave thus acts as a pump for growth of ω_s wave called the ***signal*** wave and ω_i called the ***idler*** wave. Thus, the difference frequency process is also an amplification process for the idler wave. This is called OPTICAL PARAMETERIC AMPLIFICATION (OPA). As for the laser, a constructive feedback can convert an OPA into an OPO- or an OPTICAL PARAMETERIC OSCILLATOR. As for the laser, the semi-classical treatment ignores the possibility of spontaneous generation of signal and idler pair of photons from a pump photon. Also, as for the laser, in an oscillator the signal can grow from noise which could be spontaneously generated photons or from thermal black body radiation. An OPO is thus a nonlinear optical crystal, kept in a cavity resonant at ω_s or ω_i or both, pumped by a laser at frequency $\omega_p = \omega_s + \omega_i$. Note that any pair ω_s, ω_i such that $\omega_p = \omega_s + \omega_i$ can be generated. The frequency selection comes from phase matching condition as well as the resonance condition of the oscillator. Tuning of the frequency pair ω_s, ω_i can be obtained by either rotating the crystal to change the angle between optic axis and the direction of propagation or by temperature variation which is quite effective in $LiNbO_3$ for changing the birefringence and hence the phase matched pair ω_s, ω_i.

REFERENCES:

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RECAP:

- Frequency conversion processes sum frequency generation and difference frequency generation are described by the same set of 3 coupled equations.
- overall permutation symmetry of the nonlinear susceptibility yields 2 conservation laws.
- Total power remains constant.
- The second conservation law called Manely Rowe Relations state that destrying each photon of pump wave creates one in signal wave and another in the idler wave.
- DFG process implies amplification of idler wave.
- With feed back this provides an oscillator.