

## Module 2 : Nonlinear Frequency Mixing

### Lecture 10 : Second Harmonic Generation

#### Objectives

##### In this lecture we will look at

- Detailed description of second harmonic generation.
- Coupled wave solutions.

Consider, e.g., an experiment in which a monochromatic optical field at frequency  $\omega$  is incident. In the second order, it can create  $P^{(NLS)}$  at frequencies  $2\omega$  and  $0$ . The nonlinear polarization at the second harmonic frequency is given by

$$P_{\mu}^{(NLS)}(2\omega) = \epsilon_0 \chi_{\mu\alpha\alpha}^{(2)}(-2\omega, \omega, \omega) E_{\alpha}(\omega) E_{\alpha}(\omega) \quad (10.1)$$

where summation over repeated indices is implied. Now, as  $\vec{E}(2\omega)$  becomes nonzero, it can also create a  $\vec{P}^{(NLS)}$  at  $\omega$

$$P_{\mu}^{(NLS)}(\omega) = 2\epsilon_0 \chi_{\mu\alpha\beta}^{(2)}(-\omega, 2\omega, -\omega) E_{\alpha}(2\omega) E_{\beta}^*(\omega) \quad (10.2)$$

The factor 2 accounts for the fact that there are two distinct frequencies. The two waves are therefore coupled and follow the wave equations

$$\nabla \times \nabla \times \vec{E}(2\omega) - \left(\frac{2\omega}{c}\right)^2 \vec{E}(2\omega) = \epsilon_0 \frac{(2\omega)^2}{c^2} \chi^{(2)}(-2\omega, \omega, \omega) : \vec{E}(\omega) \vec{E}(\omega) \quad (10.3)$$

$$\nabla \times \nabla \times \vec{E}(\omega) - \frac{\omega^2}{c^2} \vec{E}(\omega) = 2\epsilon_0 \frac{\omega^2}{c^2} \chi^{(2)}(-\omega, 2\omega, -\omega) : \vec{E}(2\omega) \vec{E}(\omega) \quad (10.4)$$

The coupling of the waves means that it is possible to exchange energy between them. If  $\hat{z}$  is the direction of propagation then without the nonlinearity the solution would be

$$\vec{E}(\omega) = \hat{a} A_0 e^{ikz} \quad (10.5)$$

where  $A_0$  is the amplitude and  $\hat{a}$  is the polarization. In the presence of nonlinear coupling the amplitude of the two waves can vary with  $z$ . So, we write,

$$\vec{E}(\omega) = \hat{a}_1 A_1(z) e^{ik_1 z} \quad (10.6a)$$

and,

$$\vec{E}(2\omega) = \hat{a}_2 A_2(z) e^{ik_2 z} \quad (10.6b)$$

where  $\hat{a}_1, \hat{a}_2$  are the polarization vectors such that

$$k_i^2 (\hat{z} \times \hat{z} \times \hat{a}_i) + \frac{\omega_i^2}{c^2} \vec{E}(\omega_i) \cdot \hat{a}_i = 0 \quad (10.7)$$

Thus, if  $\chi^{(2)} = 0$ ,  $A_1$  and  $A_2$  would be constants and no transfer of energy would be possible between the two waves. In the nonlinear case  $\chi^{(2)} \neq 0$ , we can assume that  $A_i(z)$  are slowly varying functions of  $z$ . i.e.

$$\frac{\partial^2 A_i}{\partial z^2} \ll k \frac{\partial A_i}{\partial z} \ll k^2 A_i \quad (10.8)$$

Substituting from eqs (10.5) and (10.6) in eqs (10.3) and (10.4) and using eq 10.7 and approximation

(10.8), we get equations for  $\partial A_1 / \partial z$  and  $\partial A_2 / \partial z$ .

Then taking scalar products of these equations with  $\hat{a}_2$  and  $\hat{a}_1$ , respectively, we obtain

$$\frac{\partial A_2}{\partial z} = \frac{i \epsilon_0 A_1^2 2 \omega^2}{c^2 k_2^2 \cos^2 \alpha_2} \tilde{\chi}^{(2)}(-2\omega, \omega, \omega) : \hat{a}_2 \hat{a}_1 \hat{a}_1 e^{-i \Delta k z} \quad (10.9)$$

and

$$\frac{\partial A_1}{\partial z} = \frac{i \epsilon_0 A_1^* A_2 \omega^2}{c^2 k_1^2 \cos^2 \alpha_1} \tilde{\chi}^{(2)}(-\omega, 2\omega, -\omega) : \hat{a}_1 \hat{a}_2 \hat{a}_1 e^{i \Delta k z} \quad (10.10)$$

where  $\Delta k = k_2 - 2k_1$  is the wave vector mismatch and  $\alpha_1$  is the angle between the electric field  $\vec{E}(\omega_1)$  and displacement vector  $\vec{D}(\omega_1)$  at the fundamental frequency and similarly for  $\alpha_2$ . The two susceptibility tensors involved here are not independent but are related to each other by the overall permutation symmetry. The susceptibility  $\chi_{\mu\alpha\beta}^{(2)}(\omega, \omega_1, \omega_2)$  is invariant under the exchange of pairs  $\mu\omega, \alpha\omega_1, \beta\omega_2$ . This can be used to define a common coupling constant for the coupled wave propagation

$$K = \epsilon_0 \tilde{\chi}^{(2)}(-2\omega, \omega, \omega) : \hat{a}_2 \hat{a}_1 \hat{a}_1 = \epsilon_0 \tilde{\chi}^{(2)*}(-\omega, 2\omega, -\omega) : \hat{a}_1 \hat{a}_2 \hat{a}_1 \quad (10.11)$$

If the conversion to the second harmonic is small, we may assume pump amplitude to remain constant. Then eq (10.9) gives:

$$\frac{\partial A_2}{\partial z} = K' e^{-i \Delta k z} \quad (10.12)$$

where the constant  $K'$  includes all factors independent of  $z$ . The solution of this is

$$A_2 = z K' e^{-(i \Delta k z / 2)} \frac{\sin(\Delta k z / 2)}{(\Delta k z / 2)} \quad (10.13)$$

where we have assumed that at  $z=0$  the second harmonic wave amplitude is zero. This shows that the second harmonic intensity  $I_2(z)$  is given by

$$I_2 \sim |A_2(z)|^2 = z^2 K'^2 \frac{\sin^2(\Delta k z / 2)}{(\Delta k z / 2)^2} \quad (10.14)$$

As anticipated earlier this is an oscillatory function. The net conversion reaches a maximum at  $z = \pi / \Delta k$  after which it starts to reduce. The length  $l_{coh} = \pi / \Delta k$  is called the coherence length. This important limiting length would be infinite if there was no dispersion. It is finite only due to the small frequency dependence of the refractive index and that small dependence determines the small distance over which effective conversion takes place. For second harmonic generation of radiation of vacuum wavelength  $\lambda$ ,  $\Delta k = 4\pi(\Delta n) / \lambda$  where  $\Delta n$  is the difference between the refractive indices of the medium at the fundamental and the harmonic frequencies. Typically for  $\Delta n$  as small as 0.01,  $l_{coh} = \pi / \Delta k$  is 25  $\mu m$  which is rather small. Oscillatory nature of the conversion efficiency is useful for obtaining  $\Delta k$  for a crystal by the "Maker Fringe" method. In this, we measure  $I_2$  as a function of crystal length either by using a wedge shaped crystal or by rotating a parallel slab crystal and determine  $\Delta k$  from the period of oscillation.

For the phase matched case ( $\Delta k = 0$ ) the solutions are not oscillatory since the energy conversion to second harmonic proceeds without a phase mismatch between the field at  $2\omega$  and the nonlinear

polarization at that frequency. In that case,

$$I_2 \sim z^2 I_1^2 \left( \chi_{eff}^{(2)} \right)^2 \quad (10.15)$$

where the effective coupling coefficient is defined by

$$\chi_{eff}^{(2)} = \sum_{i,j,k} \chi_{ijk}^{(2)} (-2\omega, \omega, \omega) a_{2i} a_{1j} a_{1k} \quad (10.16)$$

The condition  $\Delta k = 0$  called the PHASE MATCHING condition can be achieved by compensating the effect of dispersion by that of birefringence (Birefringence Phase Matching or BPM) or by periodically changing the sign of the coupling coefficient  $\chi_{eff}^{(2)}$ . These techniques are described in the next two lectures.

To summarize, the second harmonic conversion efficiency in the phase matched case is

$$\begin{aligned} \eta_{SH} &\sim (\text{crystal length})^2 \\ &\sim \text{Incident laser intensity} \\ &\sim \left( \chi_{eff}^{(2)} \right)^2 \end{aligned}$$

Thus, to have efficient conversion to the second harmonic we need crystals which have large  $\chi_{eff}^{(2)}$ , have large damage threshold so that higher intensities can be used and be available in relatively large sizes so that large lengths can be used. For efficient conversion of low power sources we need to focus the lasers to a relatively tight spot but that reduces the effective interaction length since the spot size increases rapidly due to diffraction. Exploiting guided wave propagation mitigates this problem to a great extent and this is particularly suitable for quasi phase matching. It is important to note that the effective coupling coefficient depends on the polarization vectors of the two beams relative to the crystal axes. For example, if we rotate a crystal about the propagation directions  $\chi_{eff}^{(2)}$  will change and effect  $\eta_{SH}$ .

It is remarkable that the ABDP formalism includes the effect of anisotropy of the crystal in an elegant way and also included the pump depletion effects fully. Recall that in birefringent crystals, for any given direction of propagation linear wave equation has two independent solutions. For a given  $\vec{k}, \hat{a}$  has two possible solutions one corresponding to the ordinary ray and the other corresponding to the extraordinary ray.

For the extraordinary wave  $\vec{E}$  and  $\vec{D}$  are not parallel and angle  $\alpha$  between them depends on the direction of propagation. By  $\alpha_1$  we denote the angle between  $\vec{E}(\omega)$  and  $\vec{D}(\omega)$ , and by  $\alpha_2$  that between  $\vec{E}(2\omega)$  and  $\vec{D}(2\omega)$ . Writing  $A_1 = \rho_1 e^{i\phi}$  and  $A_2 = \rho_2 e^{i\phi}$  we can separate the equations for evolution of the amplitudes and phases. It is easy to show that

$$2\rho_1^2 k_1 \cos^2 \alpha_1 / \mu_0 \omega \quad (10.17)$$

is the power flow per unit area in the  $\omega$  wave while

$$\rho_2^2 k_2 \cos^2 \alpha_2 / (\mu_0 \omega) \quad (10.18)$$

is that in the  $2\omega$  wave.

Equations of motion for  $\rho_1$  and  $\rho_2$  and the relative phase

$$\theta = \Delta k z + \phi_2(\zeta) - 2\phi_1(\zeta) \quad (10.19)$$

are obtained from those for  $A_1$  and  $A_2$ ,

$$(10.20)$$

$$\begin{aligned}\frac{d\rho_1}{dz} &= -\frac{\omega^2 K}{k_1 \cos^2 \alpha_1} \rho_1 \rho_2 \sin \theta \\ \frac{d\rho_2}{dz} &= +\frac{2\omega^2 K}{k_2 \cos^2 \alpha_2} \rho_1^2 \sin \theta\end{aligned}\quad (10.21)$$

$$\frac{d\theta}{dz} = \Delta k - 2\omega^2 K \cos \theta \left[ \frac{\rho_2}{k_1 \cos^2 \alpha_1} - \frac{\rho_1^2}{\rho_2 k_2 \cos^2 \alpha_2} \right] \quad (10.22)$$

It is important to note that the **energy transfer can occur from  $\omega$  to  $2\omega$  or  $2\omega$  to  $\omega$ , depending on the value of relative phase  $\theta$** . It does not matter, what the relative values of  $\rho_1$  and  $\rho_2$  are, so long as both of them are non zero. **This is the most important difference between coherent coupled waves and energy exchanges between incoherent systems governed by rate equations.** If  $\rho_2 = 0$ ,  $\theta_{initial}$  is arbitrary because  $\phi_2$  is arbitrary. The value of  $\phi_2$  which gives  $\sin \theta > 0$  will be adopted and  $\rho_2$  can grow from a zero value. However, if  $\rho_1 = 0$ , the non-linear coupling between the two waves vanishes for all values of the phases and the fundamental wave cannot grow back. Thus second order nonlinearity cannot generate sub harmonic of a laser if none is present to begin with. However, the sub-harmonic can grow from an arbitrarily small amplitude but the growth rate depends on this amplitude itself.

To solve these coupled non-linear equations ABDP first found two constants of motion. The remaining one equation was then integrated out. We note that the total power flowing through any  $z = \text{constant}$  surface must be same since no power is being absorbed by the medium. From eqs (10.29) and (10.30), one obtains

$$\begin{aligned}\rho_1 \frac{d\rho_1}{dz} \cdot \frac{k_1 \cos^2 \alpha_1}{\omega} + \rho_2 \frac{d\rho_2}{dz} \cdot \frac{k_2 \cos^2 \alpha_2}{2\omega} &= 0 \\ \text{or,} & \\ \left( \rho_1^2 \frac{k_1 \cos^2 \alpha_1}{\omega} + \rho_2 \frac{k_2 \cos^2 \alpha_2}{2\omega} \right) &= W = \text{constant}\end{aligned}\quad (10.23)$$

where  $W$  is the total power flowing through a unit area of any  $z = \text{constant}$  plane.

While the relative phase determines the direction of energy flow, the rate of energy flow also depends on the total power in the system and the non-linear susceptibility  $\chi_{eff}^{(2)}$  which in turn depends on the non-linear material and the polarization vectors  $\hat{a}_1$  and  $\hat{a}_2$ . To understand the behavior of coupled waves in a wide variety of situations it is instructive to work in terms of scaled distance  $\zeta$  and scaled amplitudes  $u$  and  $v$ . We put,

$$\zeta = \frac{\omega^2 K z}{k_1 \cos^2 \alpha_1} \sqrt{\frac{\omega W}{c^2 k_2 \cos^2 \alpha_2}} \quad (10.24)$$

$$u = \sqrt{\frac{k_1 c^2 \cos^2 \alpha_1}{\omega W}} \rho_1 \quad (10.25)$$

and,

$$v = \sqrt{\frac{k_2 c^2 \cos^2 \alpha_2}{\omega W}} \rho_2 \quad (10.26)$$

With these parameters eq ( 74) becomes

$$(10.27)$$

$$u^2 + v^2 = 1$$

and eqs (72) and (73) become

$$\frac{du}{d\zeta} = -uv \sin \theta \quad (10.28)$$

$$\frac{dv}{d\zeta} = u^2 \sin \theta \quad (10.29)$$

and

$$\frac{d\theta}{d\zeta} = \Delta s - \cos \theta (2v - u^2/v) \quad (10.30)$$

with

$$\Delta s = \Delta k z / \zeta \quad (10.31)$$

Here,  $u^2$  and  $v^2$  are the fractional powers in the wave at the fundamental frequency and its second harmonic, respectively.  $\Delta s$  is the scaled phase mismatch. Having incorporated the conservation of power flow in the scaling, ABDP obtained the second constant of integration by observing that using eqs (10.28) and (10.29) in eq (10.30). We may write

$$\frac{d\theta}{d\zeta} = \Delta s + \frac{\cos \theta}{\sin \theta} \frac{d}{d\zeta} (\ln u^2 v)$$

or,

$$\begin{aligned} u^2 v \sin \theta \frac{d\theta}{d\zeta} &= \Delta s u^2 v \sin \theta + \cos \theta \frac{d}{d\zeta} (u^2 v) \\ &= \Delta s v \frac{dv}{d\zeta} + \cos \theta \frac{d}{d\zeta} (u^2 v) \end{aligned} \quad (10.32)$$

$$\frac{d}{d\zeta} \left( u^2 v \cos \theta + \frac{1}{2} \Delta s v^2 \right) = 0$$

or,

$$u^2 v \cos \theta + \frac{1}{2} \Delta s v^2 = \text{constant} = \Gamma$$

Using (10.27) and (10.32) in Eq(10.28) we can obtain a non-linear equation for  $v^2$

$$\begin{aligned} v \frac{dv}{d\zeta} &= u^2 v \sin \theta = \pm \left( (u^2 v)^2 - (u^2 v \cos \theta)^2 \right)^{1/2} \\ &= \pm \left( (1 - v^2)^2 v^2 - \left( \Gamma - \frac{1}{2} \Delta s v^2 \right)^2 \right)^{1/2} \end{aligned} \quad (10.33)$$

or,

$$\frac{dv^2}{\zeta \left( v^2 (1 - v^2)^2 - \left( \Gamma - \frac{1}{2} \Delta s v^2 \right)^2 \right)^{1/2}} = \pm d\zeta$$

The denominator is a cubic in  $v^2$ . If  $v_a^2 \leq v_b^2 \leq v_c^2$  are the three roots of

$$v^2 (1-v^2)^2 + \left( \Gamma - \frac{1}{2} \Delta s v^2 \right)^2 = 0 \quad (10.34)$$

Then

$$\frac{dv^2}{\sqrt{(v^2 - v_a^2)(v^2 - v_b^2)(v^2 - v_c^2)}} = \pm 2d\zeta$$

The solution of this equation is a standard Jacobi elliptic function

$$v^2 = v_a^2 + (v_b^2 - v_a^2) \operatorname{sn}^2 \left( \sqrt{v_c^2 - v_a^2} (\zeta - \zeta_0), \gamma \right) \quad (10.35)$$

where

$$\gamma^2 = \frac{v_b^2 - v_a^2}{v_c^2 - v_a^2} \quad (10.36)$$

$\gamma$  is called the modulus. The Jacobi elliptic functions  $\operatorname{sn}$  and  $\operatorname{cn}$  are defined as follows. If,

$$u = \int_0^\psi \frac{d\psi}{\sqrt{1 - \gamma^2 \sin^2 \psi}}$$

then  $\operatorname{sn}(u, \gamma) = \sin \phi$  and  $\operatorname{cn}(u, \gamma) = \cos \phi$ . The Jacobi elliptic functions  $\operatorname{sn}$  and  $\operatorname{cn}$  are periodic function of  $u$  with period depending on  $g$ . They are easily evaluated using standard methods [Ref. See e.g. L. M. Milne-Thomson, *Handbook of Mathematical Functions*, M. Abramowitz and I. A. Stegun, Eds. New York: Dover, **1965**, ch. **16**, **17**., K.C.Rustagi, S.C.Mehendale and S.Meenakshi, IEEE J.Quant Electron, **QE18**,1029(1982), Appendix] and are available directly in computing systems like mathematica. We note the two limiting forms of the function

$$\begin{aligned} \operatorname{sn}(u, 0) &= \sin \phi \\ \text{and,} \\ \operatorname{sn}(u, 1) &= \tanh \phi \end{aligned}$$

Since  $\operatorname{sn}(u, \gamma)$  is bound by  $\pm 1$ , maximum and minimum values for  $v^2$  are given by the two roots of eq (10.34).

$$\begin{aligned} (v^2)_{\max} &= v_b^2 \\ (v^2)_{\min} &= v_a^2 \end{aligned}$$

If initially  $v = 0$  at  $\zeta = z = 0$ ,  $\Gamma = 0$ , and eq (79) becomes

$$v^2 (1-v^2)^2 - \left( \frac{1}{2} \Delta s v^2 \right)^2 = 0$$

For this case the lowest root is  $v_a^2 = 0$  and the other two roots are given by

$$v_b^2 = \left( 1 + \frac{1}{2} \left( \frac{\Delta s}{2} \right)^2 \right) \pm \sqrt{\left( 1 + \frac{1}{2} \left( \frac{\Delta s}{2} \right)^2 - 1 \right)} \quad (10.37)$$

For,  $\Delta s \gg 1$ ,  $v_b^2 = (2/\Delta s)^2$  and  $v_c^2 = (\Delta s/2)^2$  and the maximum fractional power conversion to second harmonic is  $(2/\Delta s)^2$ .

Recalling that the scaled phase mismatch  $\Delta s = \Delta k (z/\zeta)$  is the ratio between two lengths one characterizing the dispersion in the linear refractive index and the other characterizing the interaction between the two waves due to nonlinearity. The maximum power conversion is small if  $\Delta s \gg 1$  i.e. the dispersion occurs much faster than nonlinear coupling

In the phase matched case  $\Delta s = 0$  eq (44) becomes  $v_b^2 = v_c^2 = 1$  and  $v_a^2 = 0$ , with  $v_b^2 = v_c^2 = 1$ .

Then,

$$v^2 = \operatorname{sn}^2(\zeta, \gamma = 1) = \tanh^2 \zeta$$

In this case full conversion is possible to second harmonic but it takes infinite interaction length since  $\tanh \zeta \rightarrow 1$  only as  $\zeta \rightarrow \infty$ . However,  $\tanh \zeta$  is close to unity for moderate values of  $\zeta$ .

We will discuss some practical issues after describing the two techniques of phase matching in the next two lectures.

#### REFERENCES:

1. J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, Phys. Rev., **127**, 1918-1939(1962).

#### Recap

##### In this lecture we have learned about the

- Power conversion to the second harmonic is limited by mismatch between the refractive indices of the fundamental wave and the second harmonic.
- Exchange of energy between harmonic waves coupled by nonlinearity is determined by the relative phase between coupled waves.
- When phase matched near full conversion to second harmonic is possible.
- Complete analytical solutions of the coupled wave equations are given in terms of Jacobi elliptic functions which are easily evaluated.