

Module 2 : Nonlinear Frequency Mixing

Lecture 9 : Nonlinear Wave Propagation

Objectives

In this lecture we will look at

- Coupling of waves at different frequencies due to nonlinear polarization.
- Generation of new frequencies.
- Limits to energy transfer because of dispersion.

We begin with the coupled nonlinear wave propagation formulation by ABDP(Ref. from last page of Lecture 11) which is completely general in that it includes the effects of dispersion and anisotropy of materials as well as pump depletion effects. In the presence of electric dipole polarization the wave equation for the Fourier component $\vec{E}(\omega)$ of the electric field $\vec{E}(t)$ is given by

$$\vec{\nabla} \times \vec{\nabla} \times \vec{E}(\omega) - \frac{\omega^2}{c^2} \vec{E}(\omega) = \mu_0 \omega^2 \vec{P}(\omega) \quad (9.1)$$

We write

$$\vec{P}(\omega) = \vec{P}^{(L)}(\omega) + \vec{P}^{(NL)}(\omega) \quad (9.2)$$

where the first term on the right hand side is the linear polarization proportional to the field and the second term is the nonlinear polarization. We define the linear susceptibility tensor by

$$\vec{P}^{(L)}(\omega) = \epsilon_0 \vec{\chi}^{(1)}(\omega) \cdot \vec{E}(\omega) \quad (9.3)$$

Using

$$\vec{\epsilon}(\omega) = \epsilon_0 \left(\vec{1} + \vec{\chi}(\omega) \right) \quad (9.4)$$

the wave equation for the electric field at any frequency ω now becomes

$$\vec{\nabla} \times \vec{\nabla} \times \vec{E}(\omega) - \frac{\omega^2}{c^2} \vec{\epsilon}(\omega) \cdot \vec{E}(\omega) = \frac{\omega^2}{c^2} \vec{P}^{(NL)}(\omega) \quad (9.5)$$

The nonlinear polarization $\vec{P}^{(NL)}(\omega)$ at frequency ω as can be generated by the joint action of any number of fields at frequencies $\omega_1, \omega_2, \omega_3, \dots$ such that the algebraic sum of all frequencies is ω , i.e.,

$$\omega = \omega_1 + \omega_2 + \dots + \omega_n \quad (9.6)$$

Recall that we use complex amplitudes i.e., a monochromatic plane wave with the field

$$\vec{E} = \vec{E}_0 \cos(\omega t + \phi) = \vec{E}_0 e^{-i\phi} e^{-i\omega t} + \text{complex conjugate} \quad (9.7)$$

is written as

$$\vec{E}(t) = \vec{E}(\omega) e^{-i\omega t} + \vec{E}(-\omega) e^{i\omega t} \quad (9.8)$$

with

$$\vec{E}^*(\omega) = \vec{E}(-\omega) = (1/2) \vec{E}_0 e^{-i\phi} e^{-i\omega t} \quad (9.9)$$

with the provision that the frequencies ω_i can have either sign. [Note: some authors include a factor half on the right hand side of eq (9.8) which makes factors of 2^n appear in nth order susceptibilities.]

Eq. (9.5) is an inhomogeneous partial differential equation for $\vec{E}(\omega)$ whose general solution consists of two parts: (i) solution of linear homogeneous equation and (ii) the particular integral proportional to

\vec{P}^{NL} . If initially no wave is present at ω , the first part is zero and $\vec{E}(\omega) \sim \vec{P}^{NL}(\omega)$ or, in other words the **nonlinear polarization acts as a source of the field at ω** . The underlying physical picture is that dipole oscillators with a nonlinear response create polarizations oscillating at harmonic and combination frequencies which then radiate at these frequencies. So, the nonlinear polarization is called the non linear source polarization and denoted by $\vec{P}^{(NLS)}$.

Before proceeding further, let us recall that the energy stored in the polarization changes at a rate $\vec{E} \cdot \frac{\partial \vec{P}}{\partial t}$ independent of the functional form of $\vec{P}(\vec{E})$ -linear or otherwise. With this identification follows the energy conservation equation becomes [See Jackson]

$$\frac{dW_{mech}}{dt} + \int dV (\vec{H} \cdot \frac{\partial \vec{E}}{\partial t} + \epsilon_0 \vec{E} \cdot \frac{\partial \vec{E}}{\partial t} + \vec{E} \cdot \frac{\partial \vec{P}}{\partial t}) + \int \vec{S} \cdot d\vec{a} = 0 \quad (9.10)$$

where \vec{S} is the Poynting vector and W_{mech} is the mechanical energy in the medium enclosed in a volume V . This equation derived from the Lorentz Force equation and the Maxwell's field equations states that the loss of mechanical energy of the medium with the energy in the electromagnetic field represented by the next two terms and the energy stored in the polarization is the net energy flowing out from the closed volume represented by the Poynting's vector.

When the electric field and the polarization both contain several discrete frequencies, we may write

$$\vec{E}(t) = \sum_{\omega_i} \vec{E}(\omega_i) e^{-i\omega_i t} \quad (9.11)$$

and

$$\vec{P}(t) = \sum_{\omega_i} \vec{P}(\omega_i) e^{-i\omega_i t} \quad (9.12)$$

where the sum is over all frequencies including negative frequencies representing the complex conjugate terms. Then, the time average of the rate at which polarization energy changes is given by

$$\begin{aligned} \left\langle \vec{E} \frac{\partial \vec{P}}{\partial t} \right\rangle &= \sum_{\omega_i, \omega_j} \vec{E}(\omega_i) \cdot \vec{P}(\omega_j) (-i\omega_j) \left\langle e^{-i(\omega_i + \omega_j)t} \right\rangle \\ &= \sum_{\omega_i, \omega_j} \vec{E}(\omega_i) \cdot \vec{P}(\omega_j) (-i\omega_j) \delta_{\omega_i, -\omega_j} \\ &= \sum_{\omega_i} \vec{E}(\omega_i) \cdot \vec{P}^*(\omega_i) (-i\omega_i) - c.c. \\ &= -2 \sum_{\omega_i} \omega_i \text{Im}(\vec{E}(\omega_i) \cdot \vec{P}^*(\omega_i)) \end{aligned} \quad (9.13)$$

Thus we can identify $2\omega \text{Im}(\vec{E}(\omega) \cdot \vec{P}^*(\omega))$ as the **rate at which the polarization at frequency ω exchanges energy with the field at that frequency**. For wave propagation in transparent (lossless) media there is neither Joule heating nor is the total energy contained in the electromagnetic field changing. The only energy transaction is between waves at different frequencies caused by their coupling by the nonlinearity.

Now, the rate at which the polarization radiates energy in the field at frequency ω is $\sim \text{Im}(\vec{P}^{NLS}(\omega) \cdot \vec{E}^*(\omega))$ oscillates in a sinusoidal manner with period $\pi / |\vec{k}_{NLS} - \vec{k}|$ where \vec{k}_{NLS} is the propagation vector for the nonlinear polarization and \vec{k} that for the field at the frequency ω . The polarization $\vec{P}^{(NLS)}$ travels with a wave vector \vec{k}_{NLS} determined by the fields that create this polarization i.e.

$$\vec{k}_{NLS} = \vec{k}_1 + \vec{k}_2 + \dots + \vec{k}_n \quad (9.14)$$

On the other hand the field at frequency ω travels with wave vector determined by the refractive index for that wave. This difference between \vec{k}_{NLS} and \vec{k} makes the energy transfer an oscillatory function.

Only when the two wave vectors match does the energy continue to transfer from nonlinear polarization to the field at ω . This underlines the importance of the phase matching in boosting the efficiency of nonlinear frequency conversion. As we will see later, in quasi-phase matching the sign of energy flow into the field at the generated frequency is maintained positive by changing the sign of non linear source polarization $\vec{P}^{(NLS)}$ whenever $\sim \text{Im}(e^{i(\vec{k}_{NLS}-\vec{k})z})$ changes sign.

To see this more explicitly, let us consider the generation of the wave at frequency ω by a *constant* nonlinear source polarization. This means that the energy in the generated wave is *small* compared to that in the fields which create this nonlinear polarization given by

$$\begin{aligned} P_{\mu}^{(n)}(\omega) &= N_d \chi_{\mu\alpha_1...\alpha_n}^{(n)}(-\omega, \omega_1, \dots, \omega_n) E_{\alpha_1}(\omega_1) \dots E_{\alpha_n}(\omega_n) \delta_{\alpha_1, \alpha_1 + \alpha_2 + \dots + \alpha_n} \\ &= \text{constant } e^{i(\vec{k}_1 + \dots + \vec{k}_n)z} \end{aligned} \quad (9.15)$$

where the constant is proportional to $\chi^{(n)}$ and the amplitudes of all the input waves.

If \hat{z} is the direction of propagation then without the nonlinearity the solution would be

$$\vec{E}(\omega) = \hat{a} A_0 e^{i\vec{k}z} \quad (9.16)$$

where A_0 is the amplitude. A_0 is zero if this wave is not present at $z=0$. \hat{a} is the polarization and is related to the wave vector by the equation.

$$k^2 (\hat{z} \times \hat{z} \times \hat{a}) + \frac{\omega^2}{c^2} \vec{\epsilon}(\omega) \cdot \hat{a} = 0 \quad (9.17)$$

In the presence of nonlinear coupling the amplitude of the wave can vary with z . So, we write,

$$\vec{E}_i(\omega_i) = \hat{a} A_i(z) e^{i\vec{k}z} \quad (9.18)$$

where $\omega_i = \omega$ or 2ω and A_i are the corresponding amplitudes.

We also assume that $A_i(z)$ are slowly varying functions of z , i.e.

$$\frac{\partial^2 A_i}{\partial z^2} \ll k \frac{\partial A_i}{\partial z} \ll k^2 A_i \quad (9.19)$$

Substituting from eqs (9.18) in eq (9.5) and using eq (47) and approximation (9.19), we get an equations for $\partial A_i / \partial z$. Then, taking scalar products of this equation with \hat{a} , respectively, we obtain.

$$\frac{\partial A_2}{\partial z} = K' e^{-i\Delta k z} \quad (9.20)$$

where the constant K' includes all factors independent of z . $\frac{\partial A_1}{\partial z} = 0$ since we neglect pump depletion here.

The solution of eq (9.20) with

$$A_2(0) = 0$$

is

$$A = zK'e^{-(i\Delta kz/2)} \frac{\sin(\Delta kz/2)}{(\Delta kz/2)} \quad (9.21)$$

This shows that the second harmonic intensity $I(z)$ of the generated wave is given by

$$I_2 \sim |A(z)|^2 = z^2 K'^2 \frac{\sin^2(\Delta kz/2)}{(\Delta kz/2)^2} \quad (9.22)$$

Clearly, to get substantial amplitude of the generated frequency wave we need to make the phase mismatch as small as possible.

We shall now consider some specific cases of frequency conversion detail beginning with the second harmonic (SGH) in the next lecture.

Recap

In this lecture we have learned about the

- Nonlinear polarization at any frequency acts as a source of electromagnetic field at that frequency.
- It provides an important method to generate coherent radiation at new frequencies.
- substantial frequency conversion requires phase-matching i.e. making nonlinear source polarization and the generated field travel with the same phase velocities.