Lecture 3. Wave equation description of NLO

Learning objectives:

1. The wave eq. for nonlinear media
2. Coupled amplitude equations (CAE)
3. phase matching and coherent length

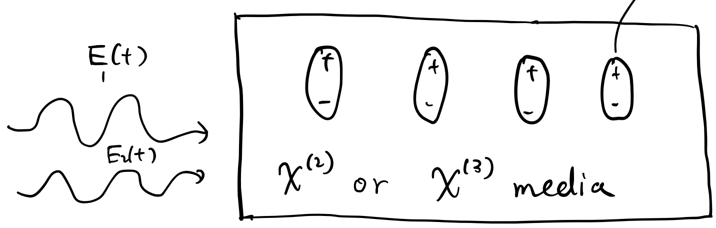
Recap:

In previous Lectures:

$$P(t) = \underbrace{P^{(1)}(t) + P^{(2)}(t) + P^{(3)}(t)}_{}$$

many freq. components.

$P(2\omega_1,\ \omega_1+\omega_2,\ \omega_1-\omega_2,\ 3\omega_1,\ 3\omega_2 \dots)$

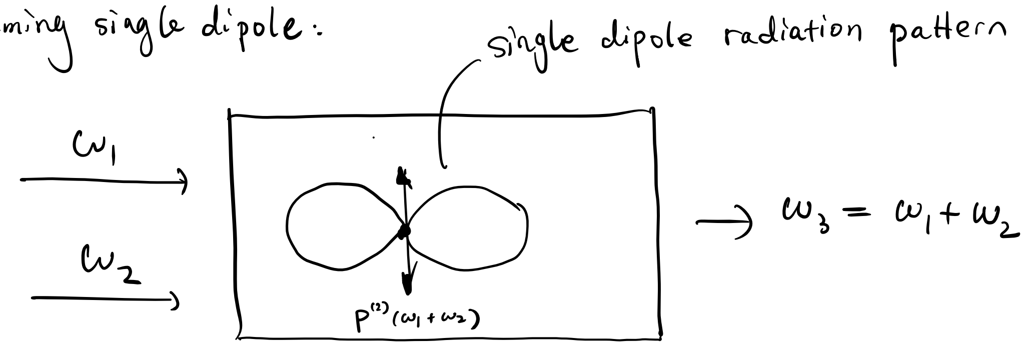


The new freq. components of the polarization act as Sources of new frequency components of the E&M field.

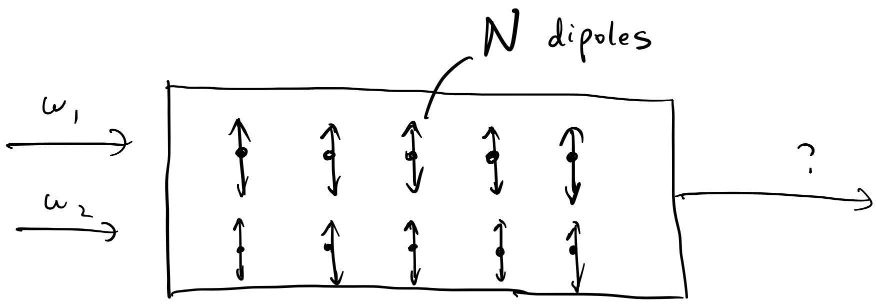
Next question: how are the EM field at other frequencies generated? Why it is not that straight forward?

An intuitive physical picture:

Assuming single dipole:



In real materials, there are N dipoles, each is oscillating with a phase that is determined by the phases of incident fields



The wave emitted by the "dipole array" can either constructively interfere or destructively interfere.

The system can act as a "phase array" of dipoles when phase matching conditions can be satisfied.

In this case: $E_0 = N \cdot E_{dipole}$. $P_0 = N \cdot P_{dipole}$.

1. The wave equation for nonlinear media

Maxwell equations:

$$\begin{cases} \nabla \cdot \vec{D} = \cancel{\rho} & ① \\ \nabla \cdot \vec{B} = 0 & ② \\ \nabla \times \vec{E} = -\dfrac{\partial \vec{B}}{\partial t} & ③ \\ \nabla \times \vec{H} = \dfrac{\partial \vec{D}}{\partial t} + \cancel{\vec{J}} & ④ \end{cases}$$

no free electron

no conducting current.

Also, $\vec{B} = \mu_0 \vec{H}$. (nonmagnetic)

$\vec{D} = \epsilon_0 \vec{E} + \vec{P}$

For linear optics $\vec{P} \sim \vec{E}$

nonlinear optics $\vec{P}$ depends nonlinearly with $\vec{E}$

$\nabla \times$ (3).

$$\nabla \times \nabla \times \vec{E} + \frac{\partial \nabla \times B}{\partial t} = 0$$

$$\nabla \times \nabla \times \vec{E} + \mu_0 \frac{\partial \nabla \times \vec{H}}{\partial t} = 0$$

$$\vec{D} = \epsilon_0 \vec{E} + \vec{P}$$

$$\nabla \times \nabla \times \vec{E} + \mu_0 \frac{\partial^2 \vec{D}}{\partial t} = 0$$

$$\Rightarrow \quad \nabla \times \nabla \times \vec{E} + \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \vec{E} = -\frac{1}{\epsilon_0 c^2} \frac{\partial^2 \vec{P}}{\partial t^2}$$

Use the identity: $\nabla \times \nabla \times \vec{E} = \nabla(\overset{\to 0}{\nabla \cdot \vec{E}}) - \nabla^2 \vec{E}$, we get

$$\nabla^2 \vec{E} - \frac{1}{\epsilon_0 c^2} \frac{\partial^2}{\partial t^2} \vec{E} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \vec{P}}{\partial t^2}$$

or $$\nabla^2 E - \frac{1}{\epsilon_0 c^2} \frac{\partial^2}{\partial t^2} \vec{D} = 0 \qquad (5)$$

Split: $\vec{P} = \vec{P}^{(1)} + \vec{P}^{NL}$

$$\vec{D} = \underbrace{\epsilon_0 \vec{E} + \vec{P}^{(1)}}_{D^{(1)}} + \vec{P}^{NL} = \vec{D}^{(1)} + \vec{P}^{NL},$$

plug into (5)

$$\nabla^2 \vec{E} - \frac{1}{\epsilon_0 c^2}\frac{\partial^2 D^{(1)}}{\partial t^2} = \frac{1}{\epsilon_0 c^2}\frac{\partial^2 P^{NL}}{\partial t^2} \quad (6)$$

relative dielectric constant

Let $D^{(1)} = \epsilon_0 \epsilon^{(1)} E$, plug in (6), we get.

$$\boxed{\nabla^2 E - \frac{\epsilon^{(1)}}{c^2}\frac{\partial^2 \vec{E}}{\partial t^2} = \frac{1}{\epsilon_0 c^2}\frac{\partial^2 \vec{P}^{NL}}{\partial t^2}} \quad (7)$$

source

Comments:

(1) Eq. (7) is the generic wave equation.

(2) The nonlinear response of the medium (P^{NL}) acts as a source term. When $P^{NL}=0$, the equation becomes the regular wave equation for linear optics. The solution is free waves propagating at a velocity $c/\sqrt{\epsilon^{(1)}}$

(3) For different frequency components.

$$\boxed{\nabla^2 E_n - \frac{\epsilon^{(1)}(\omega_n)}{c^2}\frac{\partial^2 E_n}{\partial t^2} = \frac{1}{\epsilon_0 c^2}\frac{\partial^2 P_n^{NL}}{\partial t^2}}$$

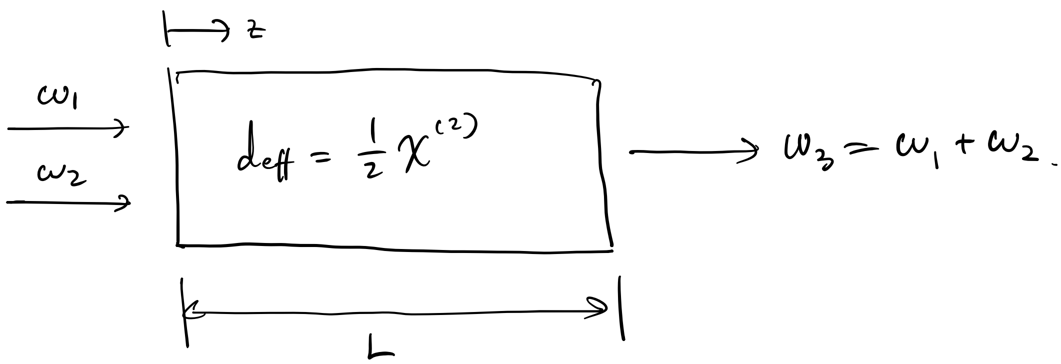
i.e. eq. (7) is valid for different freq. components.

proof: P73. Boyd.

2. Coupled amplitude equation (CAEs)

Problem to solve: use the "generic wave eq." to describe nonlinear processes.

An example: SFG in $\chi^{(2)}$ media



We expect the solution of the "generic wave eq." to be in the form:

$k_3 = n_3 \frac{\omega_3}{c}$. $n_3 = \sqrt{\epsilon^{(1)}(\omega_3)}$

$$\vec{E}_3(z,t) = \underset{\uparrow}{A_3}\, \underbrace{e^{i(k_3 z - \omega_3 t)}}_{\text{plane wave}} + c.c. \quad (1)$$

slowly varying along z.

To get this, the nonlinear source is in the form:

$$\vec{P}_3(z,t) = P_3 e^{-i\omega_3 t} + c.c.$$

$P_3 = 4\epsilon_0 d_{eff} \cdot E_1 \cdot E_2$ (last lecture)

Assuming input fields are $E_1(z,t) = A_1 e^{i(k_1 z - \omega_1 t)} + c.c.$

$E_2(z,t) = A_2 e^{i(k_2 z - \omega_2 t)} + c.c.$

So $P_3(z,t) = 4\epsilon_0 d_{eff} \cdot A_1 A_2 e^{i[(k_1+k_2)z - \omega_3 t]}$ ②

Generic wave equation:

$$\nabla^2 E_n + \frac{\omega_n^2}{c^2}\epsilon^{(1)}(\omega_n) E_n = -\frac{\omega_n^2}{\epsilon_0 c^2} P_n^{NL} \quad ③$$

Plug ①, ② into ③, we get:

equal

$$\left[\frac{d^2A_3}{dz^2} + 2ik_3\frac{dA_3}{dz} - \cancel{k_3^2 A_3} + \cancel{\frac{\epsilon^{(1)}(\omega_3)\,\omega_3^2 \cdot A_3}{c^2}}\right] e^{i(k_3 z - \cancel{\omega_3 t})} + \cancel{c.c.}$$

$$= -\frac{4 d_{eff}\,\omega_3^2}{c^2} A_1 A_2 e^{i[(k_1+k_2)z - \cancel{\omega_3 t}]} + \cancel{c.c.}$$

$$\Rightarrow \frac{d^2A_3}{dz^2} + 2ik_3\frac{dA_3}{dz} = -\frac{4 d_{eff} \cdot \omega_3^2}{c^2} A_1 A_2 e^{i(k_1+k_2-k_3)z}$$

Use the slowly varying amplitude approx.

$$\left|\frac{d^2A_3}{dz^2}\right|^2 \ll \left|k_3\frac{dA_3}{dz}\right|^2$$

we get

$$\boxed{\frac{dA_3}{dz} = \frac{2i\, d_{eff}\, \omega_3^2}{k_3 c^2} A_1 A_2 e^{i\Delta k z}}$$

$\Delta k = k_1 + k_2 - k_3$

a.k.a. wavevector, or momentum mismatch.

coupled amplitude equation

A_3 depends on A_1 and A_2

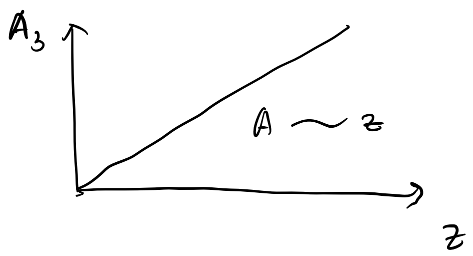
Note, A_1 and A_2 are not constants. they also vary along z

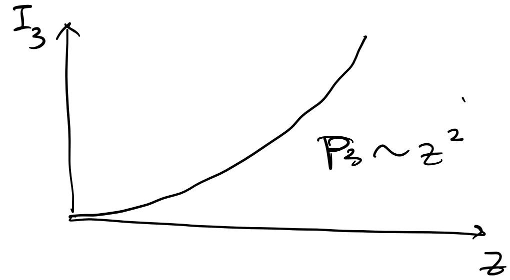
So we have a set of coupled amplitude equations:

$$\begin{cases} \dfrac{dA_3}{dz} = \dfrac{2i\, d_{eff}\, \omega_3^2}{k_1 c^2} A_1 \cdot A_2\, e^{i\Delta k z} \\[2ex] \dfrac{dA_1}{dz} = \dfrac{2i\, d_{eff}\, \omega_1^2}{k_1 c^2} A_3 A_2^* e^{-i\Delta k z} \\[2ex] \dfrac{dA_2}{dz} = \dfrac{2i\, d_{eff} \cdot \omega_2^2}{k_2 c^2} A_3 \cdot A_1^* e^{-i\Delta k z} \end{cases}$$

3. Phase matching.

① when $\Delta k = 0$. (phase matched)





Comments:

① when $\Delta k = 0$ (phase matching is satisfied), generated ω_3 maintains a fixed phase relation with the P_{NL}, and is able to extract energy most efficiently from the incident waves.

② Microscopic picture: when $\Delta k = 0$, field emitted by each dipoles add coherently in the forward direction.

② When $\Delta k \neq 0$,

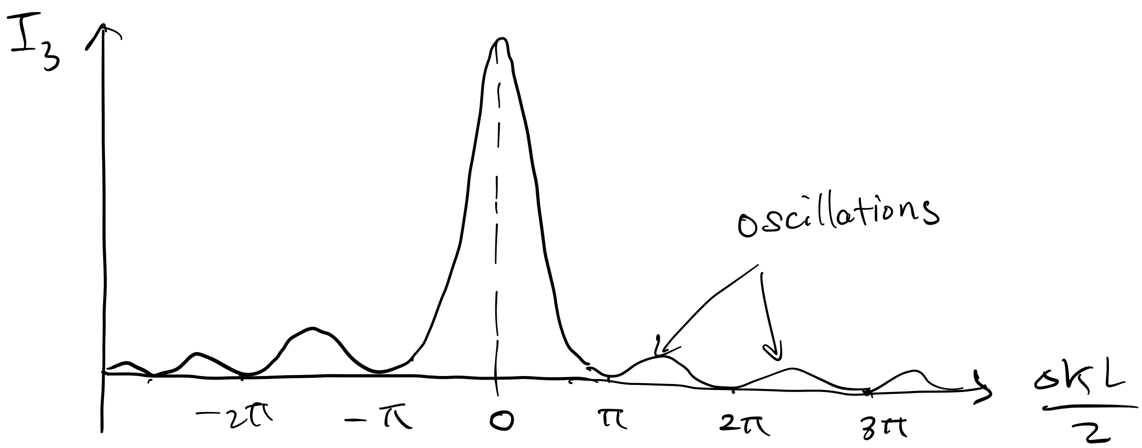
$$A_3(L) = \frac{2i\, d_{eff} \cdot \omega_3^2 \cdot A_1 A_2}{k_3 c^2} \int_0^L e^{i\Delta k z}\, dz = \frac{2i\, d_{eff}\, \omega_3^2 A_1 A_2}{k_3 c^2} \left(\frac{e^{i\Delta k L} - 1}{i\Delta k} \right)$$

Intensity of ω_3.

$$I_3 = 2 n_3 \cdot \epsilon_0 c \, |A_3|^2.$$

$$I_3 = \frac{8 n_3 \epsilon_0 d_{eff}^2 \cdot \omega_3^4 |A_1|^2 |A_2|^2}{k_3^2 c^3} \left| \frac{e^{i\Delta k L} - 1}{\Delta k} \right|^2 \rightarrow L^2 \operatorname{sinc}^2\left(\frac{\Delta k L}{2}\right)$$

$$= \frac{8 d_{eff}^2 \omega_3^2 I_1 I_2}{n_1 n_2 n_3 \epsilon_0 c^2} L^2 \operatorname{sinc}^2\left(\frac{\Delta k L}{2}\right)$$

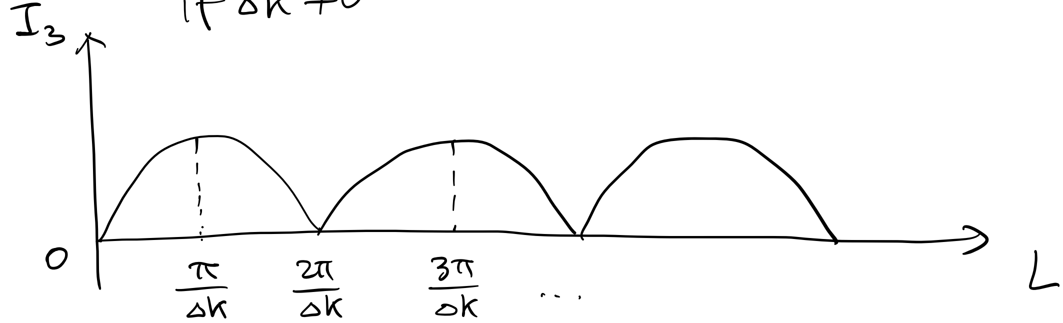


Another way:

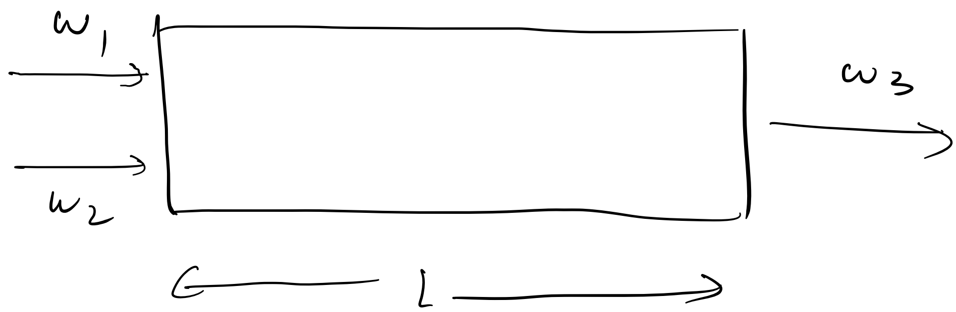
$\frac{\Delta k L}{2} = \frac{\pi}{2}$. reaches maximum

$$I_3 = I_3^{(max)} \cdot \left[\frac{\sin\left(\frac{\Delta k L}{2}\right)^2}{\left(\frac{\Delta k L}{2}\right)^2} \right]$$

if $\Delta k \neq 0$



coherent length.



Comments:

when $L < L_{coh}$, there is no phase matching issue, albeit with much lower conversion efficiency.

eg. 2D materials, Mie resonators