Lecture 5

Wave-equation description of nonlinear optical interactions; coupled-wave equations; solutions of the three-wave coupled equations.

Time-varying polarization as a source in the wave equation

Linear optics

$$P(t) = \varepsilon_0 \chi^{(1)} E(t) \qquad\qquad \chi^{(1)} = n^2 - 1$$

The formal definition of the <u>nonlinear</u> polarization:

$$\longrightarrow \quad P(t) = \varepsilon_0 \{ \chi^{(1)} E(t) + \chi^{(2)} E^2(t) + \chi^{(3)} E^3(t) + \cdots \}$$

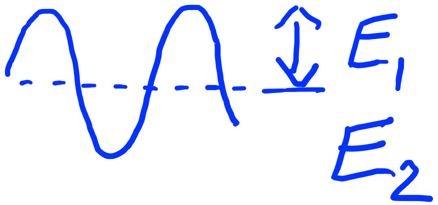
$$= P^{(1)}(t) + P^{(2)}(t) + P^{(3)}(t) + \cdots \qquad (5.1)$$

Nonlinear generation of new frequency components

Assume the optical field incident upon a <u>second-order</u> nonlinear optical $\chi^{(2)}$ medium consists of two distinct frequency components:

$$E(t) = E_1\cos(\omega_1 t) + E_2\cos(\omega_2 t) = Real\,\{E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t}\}$$

amplitudes of the frequency components at ω_1 and ω_2



From (5.1), the second-order contribution to the nonlinear polarization is of the form

$$P^{(2)}(t) = \varepsilon_0 \chi^{(2)} E(t)^2$$

Since this is a <u>nonlinear</u> relation, the optical field should be written in the <u>real</u> form:

$$E(t) = \frac{1}{2}(E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t} + E_1^* e^{-i\omega_1 t} + E_2^* e^{-i\omega_2 t}) = \frac{1}{2}E_1 e^{i\omega_1 t} + \frac{1}{2}E_2 e^{i\omega_2 t} + c.c.$$

We find that the nonlinear polarization is:

$$P^{(2)}(t) = \varepsilon_0 \chi^{(2)} \frac{1}{4}(E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t} + E_1^* e^{-i\omega_1 t} + E_2^* e^{-i\omega_2 t})^2 =$$

$$= \varepsilon_0 \chi^{(2)} \frac{1}{4}[E_1^2 e^{2i\omega_1 t} + E_2^2 e^{2i\omega_2 t} + 2E_1E_2 e^{i(\omega_1+\omega_2)t} + 2E_1E_2^* e^{i(\omega_1-\omega_2)t} + c.c.] + \varepsilon_0 \chi^{(2)} \frac{1}{2}(E_1E_1^* + E_2E_2^*)$$

SHG, second harmonic generation

SFG, sum-frequency generation

DFG, difference-frequency generation

OR, optical rectification

note that the whole expression is real !

Generation of new frequency components

In the complex representation $A\,cos(\omega t) \rightarrow \; Ae^{i\omega t}$, the amplitudes of various frequency components of the nonlinear polarization are given by:

$$at\ 2\omega_1: \qquad P(t) = \frac{1}{4}\varepsilon_0\chi^{(2)}E_1^2 e^{2i\omega_1 t} + c.c. = \frac{1}{2}\{\frac{1}{2}\varepsilon_0\chi^{(2)}\ E_1^2 e^{2i\omega_1 t} + c.c.\} = \frac{1}{2}\{P(2\omega_1)e^{2i\omega_1 t} + c.c.\}$$

$P(2\omega_1) = \frac{1}{2}\varepsilon_0\chi^{(2)}E_1^2$ <u>amplitude</u> of polarization at $2\omega_1$; can also write: $P_{2\omega_1}(t) = P(2\omega_1)\cos(2\omega_1 t)$

$$at\ 2\omega_2: \qquad P(t) = \frac{1}{4}\varepsilon_0\chi^{(2)}E_2^2 e^{2i\omega_2 t} + c.c. = \frac{1}{2}\{\frac{1}{2}\varepsilon_0\chi^{(2)}\ E_2^2 e^{2i\omega_2 t} + c.c.\} = \frac{1}{2}\{P(2\omega_2)e^{2i\omega_2 t} + c.c.\}$$

$P(2\omega_2) = \frac{1}{2}\varepsilon_0\chi^{(2)}E_2^2$ <u>amplitude</u> of polarization at $2\omega_2$; can also write: $P_{2\omega_2}(t) = P(2\omega_2)\cos(2\omega_2 t)$

Generation of new frequency components

In the complex representation $A\,cos(\omega t) \rightarrow \; Ae^{i\omega t}$, the amplitudes of various frequency components of the nonlinear polarization are given by:

polarization amplitude for...

$$P(2\omega_1) = \frac{1}{2}\varepsilon_0\chi^{(2)}E_1^2$$

SHG, second harmonic generation

$$P(2\omega_2) = \frac{1}{2}\varepsilon_0\chi^{(2)}E_2^2$$

SHG, second harmonic generation

$$P(\omega_1 + \omega_2) = \varepsilon_0\chi^{(2)}E_1E_2$$

SFG, sum-frequency generation

$$P(\omega_1 - \omega_2) = \varepsilon_0\chi^{(2)}E_1E_2^*$$

DFG, difference-frequency generation

$$P(0) = \frac{1}{2}\varepsilon_0\chi^{(2)}(E_1E_1^* + E_2\,E_2^*) = \frac{1}{2}\varepsilon_0\chi^{(2)}(|E_1|^2 + |E_2|^2)$$

OR, optical rectification

(5.2)

Generation of new frequency components

Do <u>without</u> complex representation, simply $E_1(t) = E_1 cos(\omega t)$

$$P^{(2)}(t) = \varepsilon_0 \chi^{(2)} {E_1}^2(t) = \varepsilon_0 \chi^{(2)} {E_1}^2 cos^2(\omega t) = \varepsilon_0 \chi^{(2)} {E_1}^2 \frac{1}{2}[1 + cos(2\omega t)]$$

$$P(2\omega_1) = \frac{1}{2} \varepsilon_0 \chi^{(2)} E_1^2$$

amplitude of the frequency component at $2\omega_1$

$$P(DC) = \frac{1}{2} \varepsilon_0 \chi^{(2)} E_1^2$$

DC polarization

Second Harmonic Generation

amplitude of the frequency component at $2\omega_1$

Amplitude $$P(2\omega_1) = \frac{1}{2}\varepsilon_0 \chi^{(2)} E_1^2$$

$$\boxed{d_{NL} = \frac{1}{2}\chi^{(2)}}$$

$\frac{1}{2}$ factor
– a sequence of historical convention

→ $$P(2\omega_1) = \varepsilon_0 d_{NL} E_1^2$$

in the same way

$$P(2\omega_2) = \varepsilon_0 d_{NL} E_2^2$$

Generation of new frequency components

In the complex representation $A\,cos(\omega t) \rightarrow \; Ae^{i\omega t}$, the amplitudes of various frequency components of the nonlinear polarization are given by:

$$\left.\begin{aligned} P(2\omega_1) &= \varepsilon_0 d_{NL} E_1^2 && \text{SHG, second harmonic generation} \\ P(2\omega_2) &= \varepsilon_0 d_{NL} E_2^2 && \text{SHG, second harmonic generation} \\ P(\omega_1 + \omega_2) &= 2\varepsilon_0 d_{NL} E_1 E_2 && \text{SFG, sum-frequency generation} \\ P(\omega_1 - \omega_2) &= 2\varepsilon_0 d_{NL} E_1 E_2^* && \text{DFG, difference-frequency generation} \\ P(0) &= \varepsilon_0 d_{NL}(|E_1|^2 + |E_2|^2) && \text{OR, optical rectification} \end{aligned}\right\} \quad (5.2a)$$

Coupled -wave theory (plane waves)

Recall slowly varying envelope approximation (SVEA) equation (2.3) from lecture 2

$$\boxed{\frac{\partial E(z)}{\partial z} = -\frac{i\omega c}{2n}\mu_0 P_{ext} = -\frac{i\omega}{2nc\varepsilon_0} P_{ext}} \qquad (2.11)$$

perturbation polarization

Now the role of perturbation polarization P_{ext} is played by the <u>nonlinear polarization</u> P_{NL}

Assume we have 3 interacting waves $E_1 e^{i\omega_1 t}$, $E_2 e^{i\omega_2 t}$, $E_3 e^{i\omega_3 t}$ such that $\omega_1 + \omega_2 = \omega_3$

DFG $\omega_1 = \omega_3 - \omega_2$
DFG $\omega_2 = \omega_3 - \omega_1$
SFG $\omega_3 = \omega_1 + \omega_2$

For nonlinear polarizations We can write, see (5.2a):

$$P(\omega_1) = 2\varepsilon_0 d\, E_3 E_2^*$$
$$P(\omega_2) = 2\varepsilon_0 d\, E_3 E_1^* \qquad (5.3)$$
$$P(\omega_3) = 2\varepsilon_0 d\, E_1 E_2$$

$here \quad d \equiv d_{NL}$

Coupled -wave theory

Assume that there is no absorption in the material

From (2.11) and (5.3) it follows that:

$$\frac{dE_1}{dz} = -\frac{i\omega_1 d}{n_1 c} \; E_3 E_2^* \qquad (5.4a)$$

$$\frac{dE_2}{dz} = -\frac{i\omega_2 d}{n_2 c} \; E_3 E_1^* \qquad (5.4b)$$

$$\frac{dE_3}{dz} = -\frac{i\omega_3 d}{n_3 c} \; E_1 \, E_2 \qquad (5.4c)$$

linear diff. equations, hence we are using the complex form

The three waves are travelling waves $E_1 \rightarrow E_1(z)e^{i(\omega_1 t - k_1 z)}, \; E_2 \rightarrow E_2(z)e^{i(\omega_2 t - k_2 z)} \; , E_3 \rightarrow E_3(z)e^{i(\omega_3 t - k_3 z)}$

Take for example (5.4c) While the phase of E_3 is changing as $\omega_3 t - k_3 z$

The phase of the righ side (~ $E_1 E_2$) is changing as $(\omega_1 + \omega_2)t - (k_1 + k_2)z = \omega_3 t - (k_1 + k_2)z$

Despite of the fact that $\omega_3 = \omega_1 + \omega_2$, $k_3 \neq k_1 + k_2$

$k_3 - k_2 - k_1 = \Delta k \neq 0$ phase mismatch because of wave dispersion

Three waves have different phase velocities. As a result, the induced polarization at ω_3 moves at a different velocity than the field at ω_3

$$phase\ velocity = \frac{\omega_3}{k_3} \neq \frac{\omega_1 + \omega_2}{k_1 + k_2}$$

We will look at this so-called ‘phase matching’ problem in Lecture 8

Coupled -wave theory

As a result of this mismatch of the sum of k–vectors, the term $e^{i\Delta kz}$ should be added to (5.4):

$$\Delta k = k_3 - k_2 - k_1$$

Δk > 0 for normal-dispersion material

$$\frac{dE_1}{dz} = -\frac{i\omega_1 d}{n_1 c} E_3 E_2^* e^{-i\Delta kz}$$

$$\frac{dE_2}{dz} = -\frac{i\omega_2 d}{n_2 c} E_3 E_1^* e^{-i\Delta kz} \qquad (5.5)$$

$$\frac{dE_3}{dz} = -\frac{i\omega_3 d}{n_3 c} E_1 E_2 e^{i\Delta kz}$$

$\boxed{d \equiv d_{NL} = \chi^{(2)}/2}$

Coupled -wave theory

from Lecture 2:

Energy flux (intensity): $I = \frac{1}{2}(c/n)\varepsilon|E|^2 = \frac{1}{2}cn\varepsilon_0|E|^2 = |E|^2$ /2η Watts per m^2

Photon flux: $\frac{I}{\hbar\omega} = \frac{c\varepsilon_0 n|E|^2}{2\hbar\omega} = (\frac{c\varepsilon_0}{2\hbar})\frac{n|E|^2}{\omega} \sim \frac{n}{\omega}|E|^2$ (5.6) photons per m^2 per second

introduce a new field variable : $A = \sqrt{\frac{n}{\omega}}E$ (5.7)

such that $|A|^2$ is now proportional to the <u>photon flux:</u> $\Phi = \frac{c\varepsilon_0}{2\hbar}|A|^2$ -photons per m^2 per sec

Intensity: $I = \frac{1}{2}cn\varepsilon_0|E|^2 = \frac{1}{2}cn\varepsilon_0(\frac{\omega}{n})|A|^2 = \frac{c\varepsilon_0}{2}\omega|A|^2 \sim \omega|A|^2$

Coupled -wave theory

Now rewrite (5.5): $\boxed{E \to \sqrt{\frac{\omega}{n}} A}$

$$\sqrt{\frac{\omega_1}{n_1}} \frac{dA_1}{dz} = -\frac{i\omega_1 d}{n_1 c} \sqrt{\frac{\omega_3}{n_3}} \sqrt{\frac{\omega_2}{n_2}} A_3 A_2^* e^{-i\Delta kz}$$

$$\sqrt{\frac{\omega_2}{n_2}} \frac{dA_2}{dz} = -\frac{i\omega_2 d}{n_2 c} \sqrt{\frac{\omega_3}{n_3}} \sqrt{\frac{\omega_1}{n_1}} A_3 A_1^* e^{-i\Delta kz}$$

$$\sqrt{\frac{\omega_3}{n_3}} \frac{dA_3}{dz} = -\frac{i\omega_3 d}{n_3 c} \sqrt{\frac{\omega_1}{n_1}} \sqrt{\frac{\omega_2}{n_2}} A_1 A_2 e^{i\Delta kz}$$

and get:

$$\frac{dA_1}{dz} = -i \frac{d}{c} \sqrt{\frac{\omega_1 \omega_2 \omega_3}{n_1 n_2 n_3}} A_3 A_2^* e^{-i\Delta kz}$$

$$\frac{dA_2}{dz} = -i \frac{d}{c} \sqrt{\frac{\omega_1 \omega_2 \omega_3}{n_1 n_2 n_3}} A_3 A_1^* e^{-i\Delta kz} \qquad (5.8)$$

$$\frac{dA_3}{dz} = -i \frac{d}{c} \sqrt{\frac{\omega_1 \omega_2 \omega_3}{n_1 n_2 n_3}} A_1 A_2 e^{i\Delta kz}$$

Coupled-wave theory

Define : $g = \frac{d}{c}\sqrt{\frac{\omega_1\omega_2\omega_3}{n_1 n_2 n_3}}$

g - NL coupling coefficient

$$\boxed{\begin{aligned} \frac{dA_1}{dz} &= -ig\, A_3 A_2^* e^{-i\Delta kz} \\ \frac{dA_2}{dz} &= -ig\, A_3 A_1^* e^{-i\Delta kz} \\ \frac{dA_3}{dz} &= -ig A_1 A_2 e^{i\Delta kz} \end{aligned}} \qquad (5.9)$$

This is the final form of coupled equations for 3 waves

Coupled -wave theory

Now let us find how photon fluxes at ω_1 ω_2 ω_3 are related to each other

photon flux $\sim |A|^2$

$$\frac{d}{dz}|A|^2 = \frac{d}{dz}(AA^*) = A^*\frac{dA}{dz} + A\frac{dA^*}{dz} = A^*\frac{dA}{dz} + c.c.$$

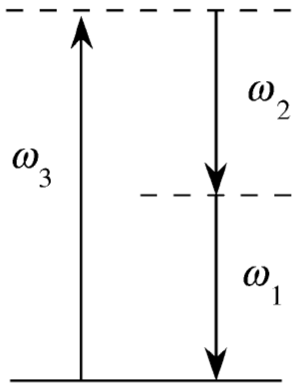
From the previous Eq. (5.7)

$$A_1^* \quad \frac{dA_1}{dz} = -ig\, A_3 A_2^* e^{-i\Delta kz} \longrightarrow \frac{d}{dz}|A_1|^2 = -ig\, A_3 A_2^* A_1^* e^{-i\Delta kz} + c.c.$$

$$A_2^* \quad \frac{dA_2}{dz} = -ig\, A_3 A_1^* e^{-i\Delta kz} \longrightarrow \frac{d}{dz}|A_2|^2 = -ig\, A_3 A_2^* A_1^* e^{-i\Delta kz} + c.c.$$

$$A_3^* \quad \frac{dA_3}{dz} = -ig A_1 A_2 e^{i\Delta kz} \longrightarrow \frac{d}{dz}|A_3|^2 = -ig A_3^* A_2 A_1 e^{i\Delta kz} + c.c. = ig\, A_3 A_2^* A_1^* e^{-i\Delta kz} + c.c.$$

Manley–Rowe relation



hence $$\boxed{\frac{d}{dz}\,|A_1|^2 = \frac{d}{dz}\,|A_2|^2 = -\,\frac{d}{dz}\,|A_3|^2} \qquad (5.8)$$ Manley–Rowe relation

same as $$\boxed{\frac{d}{dz}n_1 = \frac{d}{dz}n_2 = -\,\frac{d}{dz}n_3} \qquad (5.9)$$ *n* - number of photons

since $|A|^2 \sim \dfrac{I}{\omega}$ $$\boxed{\frac{d}{dz}\left(\frac{I_2}{\omega_2}+\frac{I_3}{\omega_3}\right)=0, \qquad \frac{d}{dz}\left(\frac{I_1}{\omega_1}+\frac{I_3}{\omega_3}\right)=0, \qquad \frac{d}{dz}\left(\frac{I_1}{\omega_1}-\frac{I_2}{\omega_2}\right)=0.} \qquad (5.10)$$

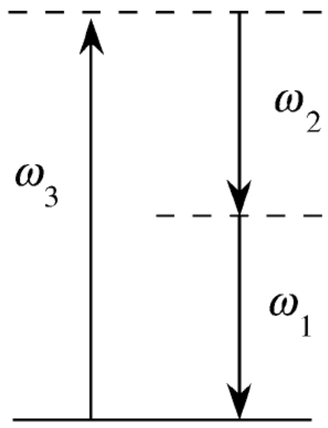
Also, using (5.10) and $\omega_1+\omega_2=\omega_3$

$$\frac{d}{dz}\,(I_1+I_2+I_3) = -\frac{\omega_1}{\omega_3}\frac{d}{dz}\,I_3 - \frac{\omega_2}{\omega_3}\frac{d}{dz}\,I_3 + \frac{d}{dz}\,I_3 = -\frac{\omega_1+\omega_2}{\omega_3}\frac{d}{dz}\,I_3 + \frac{d}{dz}\,I_3 = 0$$

$$\boxed{\frac{d}{dz}\,(I_1+I_2+I_3) = 0} \qquad (5.11)$$ energy conservation

Manley–Rowe relation

These important relations (5.8- 5.11) are universal in the sense that there may be or may be no phase matching; and also in the sense that the process can go both ways:



$$\omega_1 + \omega_2 \rightarrow \omega_3$$

or

$$\omega_3 \rightarrow \omega_1 + \omega_2$$

Of course this is under the assumption that there is no absorption in the material