Nonlinear Optics

Part I

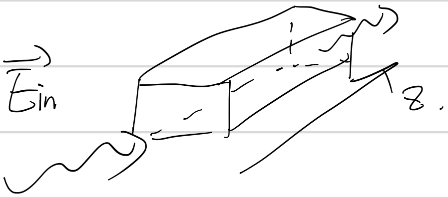
1. Second harmonic generation
2. phase matching condition/method.
3. Sum/Difference frequency generation
4. Parametric oscillation
5. Electro-optics effect (EO-modulator)
6. Nonlinear susceptility

Part II.

1. Four-wave mixing
2. Kerr nonlinearity (ultrafast laser)
3. Raman Scattering
4. Brillouin Scattering

Review of General equation of nonlinear optics

Model



Assume we know $\vec{E}(z=0) = \vec{E}_{in}$;
calculate $\vec{E}(z=L) = \vec{E}_{out}$.

First, in general, the input light could have multiple frequency components.

$$\vec{E}(z=0) = \sum_j \vec{e}_j E_j(z=0) e^{-i\omega_j t + i k_j z} + c.c.$$

Notice: E_j is not always real. In general, it is a complex quantity when more than one frequency components exist!

When you can express all E_j in real quantity, we say it's mode-locked.
Will talk about this later.

Recall our nonlinear equation of Maxwell equation:

$$\nabla^2 \vec{E} - \frac{1}{c^2}\frac{\partial^2 \vec{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \vec{P}}{\partial t^2}$$

$$\vec{P} = \vec{P}_{LN} + \vec{P}_{NL} = \varepsilon_0 \chi \vec{E} + \vec{P}_{NL}$$

$$\Rightarrow \quad \nabla^2 \vec{E} - \frac{\varepsilon_r}{c^2}\frac{\partial^2 \vec{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \vec{P}_{NL}}{\partial t^2} \quad ; \quad \varepsilon_r = n^2$$

The left-hand side is "linear".
There's no "interaction" between different frequency components.

Because differential and sum commutes. You can change their order.

$$\left(\sum_j \frac{\partial}{\partial t} a_j = \frac{\partial}{\partial t} \sum_j a_j . \right)$$

So on the left-hand side, we have:

$$\sum_j \left(\nabla^2 \vec{E}_j e^{-i\omega_j t + ik_j z} - \frac{\varepsilon_r}{c^2} \frac{\partial^2}{\partial t^2} \vec{E}_j e^{-i\omega_j t + ik_j z} + c.c. \right) = LHS.$$

Previously, we have already shown that:

$$\vec{e}_j \nabla^2 E_j(z) e^{-i\omega_j t + ik_j z} = \vec{e}_j \frac{\partial^2}{\partial z^2} \left\{ E_j(z) e^{-i\omega_j t + ik_j z} \right\}$$

$$= \vec{e}_j \left\{ \frac{\partial^2 E_j(z)}{\partial z} + 2ik_j \frac{\partial}{\partial z} E_j(z) - k_j^2 E_j(z) \right\} e^{-i\omega_j t + ik_j z} .$$

Slow varying amplitude approximation:

$$\frac{\partial^2}{\partial z^2} E_j(z) << 2ik_j \frac{\partial}{\partial z} E_j(z).$$

$$LHS: \sum_j \vec{e}_j \left(2ik_j \frac{\partial}{\partial z} E_j(z) - k_j^2 E_j(z) + \frac{\varepsilon_r \omega_j^2}{c^2} E_j(z) \right) e^{-i\omega_j t + ik_j z} + c.c.$$

Use the fact that $k_j = \frac{2\pi n_j}{\lambda_j} = \frac{n_j \omega_j}{c}$

$$-k_j^2 + \frac{\varepsilon_r \omega_j^2}{c^2} = -\frac{n_j^2 \omega_j^2}{c^2} + \frac{n_j^2 \omega_j^2}{c^2} = 0.$$

$$\text{LHS:} = \sum_j \vec{e}_j \cdot 2ik_j \frac{\partial E_j(z)}{\partial z} e^{-i\omega_j t + ik_j z} + c.c.$$

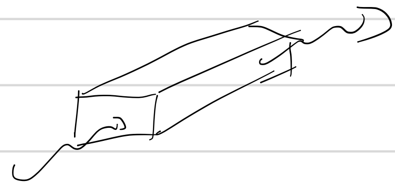
$$= \mu_0 \frac{\partial^2 \vec{P}_{NL}}{\partial t^2}$$

And $\vec{P}_{NL} = \varepsilon_0 \vec{\vec{\chi}}^{(2)} \vec{E}\vec{E} + \varepsilon_0 \vec{\vec{\chi}}^{(3)} \vdots \vec{E}\vec{E}\vec{E} + \ldots$

This is the equation you would use for the rest of the class

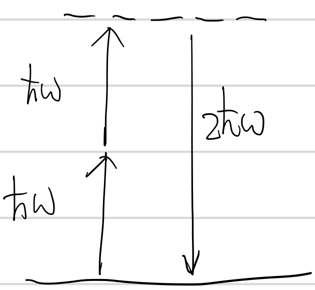
1. Second Harmonic generation

For second harmonic generation, we have:



$$P_{NL} = \varepsilon_0 \overleftrightarrow{\chi}^{(2)} \cdot \vec{E}\vec{E}$$

Correspond to physics process:



We will later calculate $\chi^{(2)}$.

From previous chapter, we know we can expand $\vec{E}$ into

$$\vec{E} = \vec{E}_1 e^{-i\omega t + ik_1 z} + \vec{E}_2 e^{-2i\omega t + ik_2 z} + c.c.$$

Question we want to answer:
If the input power is P_0, what is the output second-harmonic power? (Assume we know all parameters)

Left-hand side of the general nonlinear equation is clear:

$$LHS: \vec{e}_1 2ik_1 \frac{\partial E_1(z)}{\partial z} e^{-i\omega t + ik_1 z} + \vec{e}_2 2ik_2 \frac{\partial E_2(z)}{\partial z} e^{-2i\omega t + ik_2 z} + c.c.$$

Right-hand side: $$= \mu_0 \frac{\partial^2 \vec{P}_{NL}}{\partial t^2} = \mu_0 \varepsilon_0 \overleftrightarrow{\chi}^{(2)} \frac{\partial^2}{\partial t^2} \vec{E}\vec{E}$$

For now, for simplicity, we assume $\vec{e_1} = \vec{e_2}$ (not true in most cases)

We can simplify $\overrightarrow{\overrightarrow{\overrightarrow{\chi^{(2)}}}} \cdot \vec{E}\vec{E}$ to $\chi^{(2)} (\vec{E})^2 \vec{e}$

$$\vec{E}^2 = (E_1 + E_2 + E_1^* + E_2^*)(E_1 + E_2 + E_1^* + E_2^*)$$

In nonlinear calculation, must include c.c. all the time.

Why? $(a+a^*)^2 = a^2 + a^{*2} + 2aa^* \neq a^2 + c.c.$

$$= (E_1^2 + E_2^2 + 2E_1E_2 + c.c.)$$
$$+ 2|E_1|^2 + 2|E_2|^2$$
$$+ 2E_1E_2^* + 2E_1^*E_2$$

Notice $|E_1|^2$ is not time dependent (doesn't have $e^{-i\omega t}$)
so vanish after $\frac{\partial^2}{\partial t^2} P_{NL}$

$$\text{RHS:} = \varepsilon_0 \mu_0 \frac{\partial^2}{\partial t^2} \Big(E_1^2 e^{-2i\omega t + 2ik_1 z} + E_2^2 e^{-4i\omega t + 2ik_2 z} + 2E_1E_2 e^{-i3\omega t + i(k_1+k_2)z}$$
$$+ 2E_1E_2^* e^{+i\omega t + i(k_1-k_2)z} + c.c. \Big)$$

We will use this equation later.

$$\text{RHS:} = -\varepsilon_0 \mu_0 \omega^2 \Big(4E_1^2 e^{-2i\omega t + 2ik_1 z} + 16E_2^2 e^{-4i\omega t + 2ik_2 z} + 18E_1E_2 e^{-3i\omega t + i(k_1+k_2)z}$$
$$+ 2E_1E_2^* e^{+i\omega t + i(k_1-k_2)z} + c.c. \Big)$$

Usually in second-harmonic generation, we can assume:

$E_1(z) \gg E_2(z)$. : in low-conversion efficiency regime

A check to see if this equation make sense:

When left-handside equals right-hand side, we have:

$$2ik_1 \frac{\partial E_1(z)}{\partial z} e^{-i\omega t + ik_1 z} = -2\varepsilon_0 \mu_0 \chi_{(2)} \omega^2 E_1^* E_2 e^{-i\omega t + i(k_2 - k_1)z}$$

$$2ik_2 \frac{\partial E_2(z)}{\partial z} e^{-2i\omega t + 2ik_2 z} = -4\varepsilon_0 \mu_0 \omega^2 \chi_{(2)} E_1^2 e^{-2i\omega t + 2ik_1 z}$$

How to check equations?

Energy conservation: we are converting energy from E_1 to E_2

As we know $P = \frac{1}{2} n c \varepsilon_0 A \cdot |E|^2$.

So: $\frac{dP_1}{dz} = -\frac{dP_2}{dz}$ such that: $\frac{d}{dz}(P_1 + P_2) = 0$.

Let's work on the simplest case: $n_1 = n_2$ (refraction index is the same, normally this is not true)

then $k_2 = 2k_1$ (wavelength) ; let $k_1 = k_0 = \frac{n \cdot \omega}{c}$

So:
$$\frac{\partial E_1(z)}{\partial z} = i \frac{\varepsilon_0 \mu_0 \omega^2 \chi^{(2)}}{k_0} E_1^* E_2$$

$$\frac{\partial E_2(z)}{\partial z} = i \frac{\varepsilon_0 \mu_0 \omega^2 \chi^{(2)}}{k_0} E_1^2$$

$$\frac{\partial E_1}{\partial z} = i g E_1^* E_2 \qquad (1)$$

$$\frac{\partial E_2}{\partial z} = i g E_1^2 \qquad (2)$$

Construct $\frac{\partial}{\partial z} |E_1|^2$

$$\{ E_1^* \cdot eq.(1) + E_1 \cdot eq.(1)^* \}$$

$$= E_1^* \frac{\partial E_1}{\partial z} + E_1 \frac{\partial E_1^*}{\partial z} = \frac{\partial}{\partial z} (E_1 \cdot E_1^*) = \frac{\partial}{\partial z} |E_1|^2$$

$$= i g E_1^{*2} E_2 - i g E_1^2 E_2^*$$

Same way to construct $\frac{\partial}{\partial z} |E_2|^2$

$$E_2^* \cdot eq.(2) + E_2 \cdot eq.(2)^*$$

$$= \frac{\partial}{\partial z} |E_2|^2 = i g E_1^2 E_2^* - i g E_1^{*2} \cdot E_2$$

$$\therefore \frac{\partial}{\partial z} |E_1|^2 = i g E_1^{*2} E_2 - i g E_1^2 E_2^* = - \frac{\partial}{\partial z} |E_2|^2 .$$

Conserve energy.

(Possible problem in final exam: show energy conservation when $n_1 \neq n_2$)

Usually in second-harmonic generation, we can assume:

$E_1(z) \gg E_2(z)$: in low-conversion efficiency regime

So drop all $E_2(z)$ term.

$$\text{RHS}: = -4\mu_0\varepsilon_0\omega^2\chi E_1^2 e^{-2i\omega t+2ik_1 z} + c.c.$$

$$\text{LHS}: = 2ik_1 \frac{\partial E_1(z)}{\partial z} e^{-i\omega t+ik_1 z} + 2ik_2 \frac{\partial E_2(z)}{\partial z} e^{-2i\omega t+ik_2 z} + c.c.$$

What this means?

$$\frac{\partial E_1(z)}{\partial z} = 0. \quad \text{Non-depletion regime.} \quad ; \quad E_1(z) = E_1(0)$$

$\boxed{\text{Possible in Final exam : solve depletion regime.}}$

In non-depletion regime, we have:

$$2ik_2 \frac{\partial E_2(z)}{\partial z} e^{-2i\omega t+ik_2 z} = \chi^{(2)} 4\mu_0\varepsilon_0\omega^2 E_1^2 e^{-2i\omega t+2ik_1 z}$$

$$\Rightarrow \frac{\partial E_2(z)}{\partial z} = \frac{2i\mu_0\varepsilon_0\omega^2\chi^{(2)}}{k_2} E_1^2(z=0)\, e^{i(2k_1-k_2)z} \quad ; \quad \int e^{\alpha z} dz = \frac{e^{\alpha z}}{\alpha}$$

$$\text{Solution}: \quad E_2(z) = \frac{2i\mu_0\varepsilon_0\omega^2 E_1^2(0)\chi^{(2)}}{k_2} \int_0^L e^{i(2k_1-k_2)z} dz$$

$$= \frac{2i\mu_0\varepsilon_0\omega^2 E_1^2(0)\chi^{(2)}}{k_2} \frac{\left(e^{i(2k_1-k_2)L} - 1\right)}{i(2k_1-k_2)}$$

$$= \frac{2\mu_0\varepsilon_0\omega^2 E_1^2(0)\chi^{(2)}\left(e^{i(2k_1-k_2)L} - 1\right)}{k_2(2k_1-k_2)}$$

This is a sinc function.

First of all, $E_2(z)$ is periodic with L.
The periodicy is $(2k_1-k_2)L = 2\pi$.

So longer waveguide doesn't mean higher output.

Second: apparently, if $2k_1-k_2$ is infinitely small, there's an infitite long L to allow

$E_2 \to \infty$.

So it's possible to make the conversion from $\omega \to 2\omega$ very efficient.

Let's see what we need to do:

first, simplify the result by using $k_j = \frac{n_j \omega_j}{c}$; and $\mu_0 \varepsilon_0 = \frac{1}{c^2}$

$$E_2(z) = \frac{2\omega^2}{c^2} \frac{\chi^{(2)} E_1^2 \left(e^{i(2k_1-k_2)L} - 1\right)}{\frac{n_2 2\omega}{c}\left(\frac{2n_1\omega}{c} - \frac{2n_2\omega}{c}\right)}$$

$$= \frac{\chi^{(2)} E_1^2 \left(e^{i(2k_1-k_2)L} - 1\right)}{2(n_1-n_2)\cdot n_2}$$

Check dimension: $P = \varepsilon_0 \chi \vec{E} + \varepsilon_0 \chi^{(2)} \vec{E}^2$

$\therefore \chi^{(2)} E^2 \sim \chi E \sim E$. Dimension is correct.

$$E_2(z)\Big|_{z=L} = \frac{\chi_2 E_1^2(z=0)}{2n_2\cdot\Delta n}\left(e^{i(2k_1-k_2)L}-1\right).$$

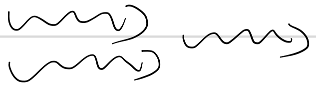
Apparently, small Δn is always good.

This is called phase-matching condition.

$\Delta n = 0$, or $\Delta k = 2k_1 - k_2 = 0$

It corresponds to momentum conservation of photon: $\hbar k$.

$2\hbar k_1$ $\hbar k_2$



We go one step further. Assume $\Delta n \neq 0$, but we have

$$\mathrm{Max}\left| e^{i(2k_1-k_2)L} - 1 \right| = 2 \quad ; \quad (2k_1-k_2)L = (2N+1)\pi.$$

$$E_2(L) = \frac{\chi_2 E_1^2}{n_2\cdot\Delta n}$$

But we usually measure power instead of E.

$P = \frac{1}{2}n\varepsilon_0 c\cdot A\cdot|E|^2$; A is mode area. Cross-section of light beam.

$$P_2 = \frac{1}{2}n_2\varepsilon_0 c\cdot A\cdot\frac{\chi_2^2}{n_2^2\cdot\Delta n^2}\cdot|E_1|^4$$

$$P_2 = \frac{1}{2} n_2 \varepsilon_0 c \cdot A \cdot \frac{\chi_2^2}{n_2^2 \cdot \Delta n^2} \cdot |E_1|^4$$

$$P_1 = \frac{1}{2} n_1 \varepsilon_0 c A \cdot |E_1|^2$$

$$\therefore \quad |E_1|^4 = \frac{4 P_1^2}{n_1^2 \varepsilon_0^2 c^2 A^2}$$

$$\Rightarrow \quad P_2 = \frac{1}{2} \frac{\varepsilon_0 c A \chi_2^2}{n_2 \cdot \Delta n^2} \cdot \frac{4 P_1^2}{n_1^2 \varepsilon_0^2 c^2 A^2}$$

$$= \frac{2 \chi_2^2 P_1^2}{n_2 n_1^2 \Delta n^2 \varepsilon_0 c \cdot A}.$$

So to increase output: 1. reduce Δn (phase-matching condition)

2. Pick large χ_2 material.

3. $P_2 \propto P_1^2$; so increase input by 2 will ↑ output by 4.

4. Reduce mode area: A of free space > A of fiber
> A of waveguide

free-space: often mm^2 : 10^{-6} m^2
fiber : $10 \mu m^2$: 10^{-10} m^2
waveguide : $1 \mu m^2$: 10^{-12} m^2.

Huge difference.