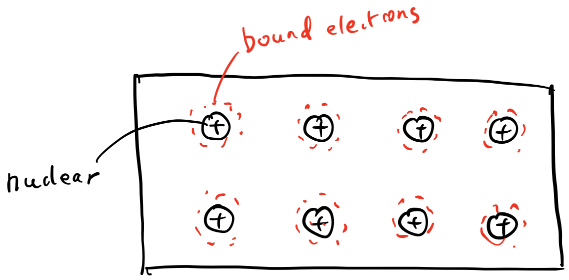
Lecture 1 Nonlinear optical processes

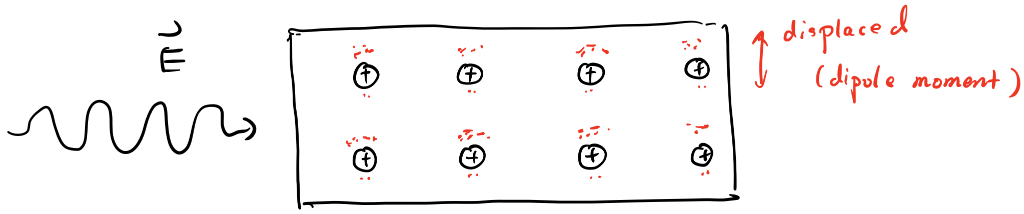
Learning objectives:

1. Review of linear optics
2. Origin of optical nonlinearity
3. Second order nonlinear processes
4. Third order nonlinear processes
5. NLO Zoology

1. Review of linear optics

Consider light incident on a material (dielectric, metal)



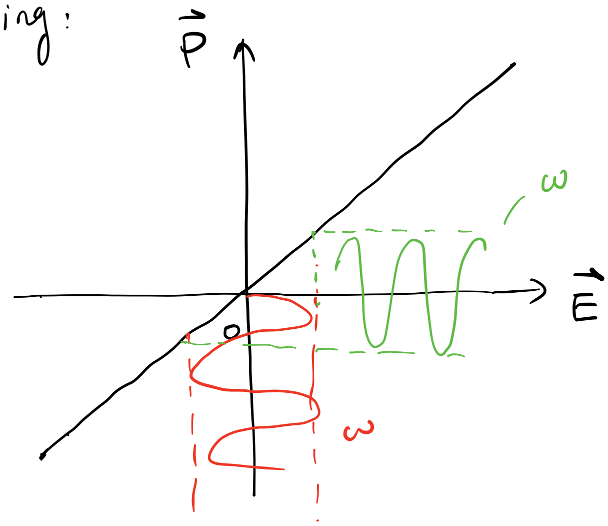


The dipole moment per unit volume :

$$\boxed{P(t) = \epsilon_0 \chi^{(1)} E(t)} \quad (1)$$

linear susceptibility .

If $E = A\cos\omega t$, $P = \epsilon_0 \chi^{(1)} \cdot A\cos\omega t$, Physical meaning:

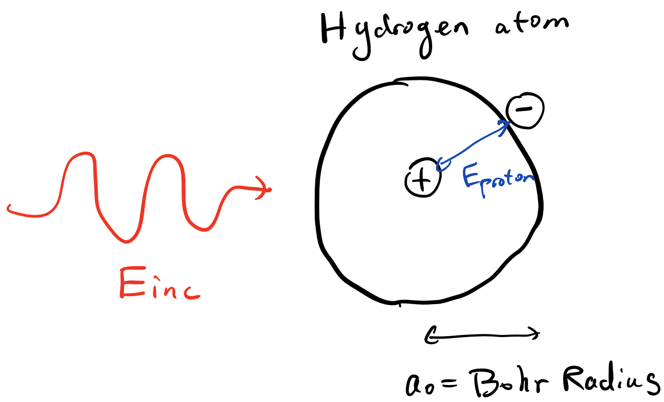


No frequency conversion!

Comments:

Equation 1 is a good approximation since in most cases, the E-field of light is much weaker than the field inside the atoms and molecules.

Consider EM wave incident on a hydrogen atom:



Forces applied to the electron:

$$F_{electron}^{EM\ wave} = q\, E_{inc}$$

$$F_{electron}^{proton} = q\, E_{proton}$$

$$E_{proton} = \frac{1}{4\pi\epsilon_0}\,\frac{q}{a_0^2} \simeq 5\times10^{11}\ V/m$$

How about E_{inc}?

For typical green laser, $P = 5mW$

$$\text{Intensity} \simeq \frac{|E_{inc}|^2}{2Z_0}$$

beam width

Assuming a beam radius of 1mm,

$$\text{Intensity} = \frac{5\,mW}{\pi \cdot 1\,mm^2} \cong 10^3 \frac{W}{m^2} = \frac{|E_{inc}|^2}{2 Z_0}$$

$$\Rightarrow \boxed{E_{inc} \simeq 10^3\ V/m \ll E_{proton}}$$

2. Origin of optical nonlinearity

Linear optics: $\tilde{p}(t) = \epsilon_0 \chi^{(1)} \tilde{E}(t)$ at low E-field.

(Harmonic oscillator model)

Generally, if consider anharmonicity,

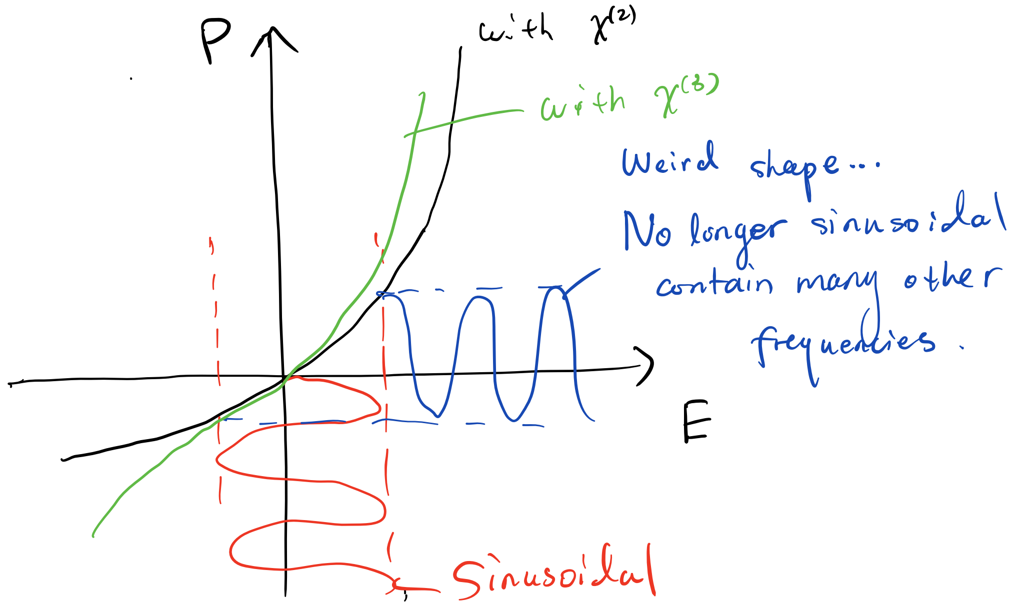
$$\tilde{P}(t) = \epsilon_0 \Big[\underbrace{\chi^{(1)} \tilde{E}(t)}_{\text{linear}} + \underbrace{\chi^{(2)} \tilde{E}(t)^2}_{\substack{\text{second order} \\ \text{(tensor)}}} + \underbrace{\chi^{(3)} \tilde{E}(t)^3}_{\substack{\text{third order} \\ \text{(tensor)}}} + \cdots \Big]$$

When $\tilde{E}(t)$ is sufficiently high, $\chi^{(2)}$, $\chi^{(3)}$ cannot be ignored.

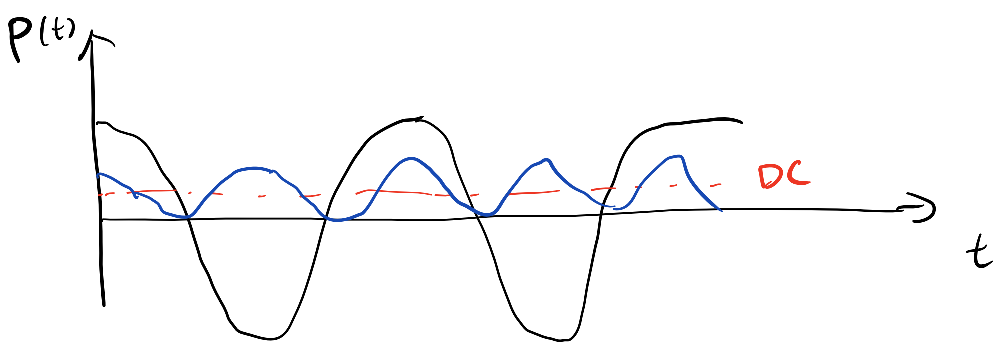
Note:

1. $\chi^{(2)}$ only occurs in non-centrosymmetric materials (no inversion symmetry)
2. $\chi^{(3)}$ can occur in all materials

In this case:



Use $\chi^{(2)}$ as an example



$$P = \epsilon_0 (\chi^{(1)} E + \chi^{(2)} E^2 + \cdots)$$

$$= \epsilon_0 (\chi^{(1)} \cdot A \cos \omega t + \chi^{(2)} \cdot A^2 \cos^2(\omega t))$$

$$= \epsilon_0 (\chi^{(1)} A \cos \omega t + \frac{1}{2} \chi^{(2)} A^2 (1 + \cos 2\omega t))$$

$$= \epsilon_0 \Big(\underbrace{\chi^{(1)} A \cos \omega t}_{\text{linear term}} + \underbrace{\tfrac{1}{2} \chi^{(2)} A^2}_{\text{DC}} + \underbrace{\tfrac{1}{2} \chi^{(2)} \cos 2\omega t}_{\text{nonlinear Term}} \Big)$$

How large are the $\chi^{(2)}$ and $\chi^{(3)}$ succeptibility ?

$$P^{(1)} = \epsilon_0 \chi^{(1)} E.$$

$$P^{(2)} = \epsilon_0 \chi^{(2)} E^2$$

$$P^{(3)} = \epsilon_0 \chi^{(3)} E^3$$

When molecules see $E_{inc} \simeq E_{proton}$, $P^{(2)} \simeq P^{(1)}$

$$\Rightarrow \chi^{(2)} \sim \frac{\chi^{(1)}}{E_{proton}} = \frac{\text{unity}}{5 \times 10^{11}\ V/m}$$

$$\simeq 2 \times 10^{-12}\ m/V$$

when $E_{inc}^2 \sim E_{proton}$, $P^{(3)} \sim P^{(1)}$

$$\Rightarrow \chi^{(3)} \sim \frac{\chi^{(1)}}{E_{proton}} = \frac{\text{unity}}{(5 \times 10^{11}\ V/m)^2} \simeq 3.78 \times 10^{-24}\ m^2/V^2$$

3. Second Order nonlinear optical processes

① Second harmonic generation (SHG)

Input field:

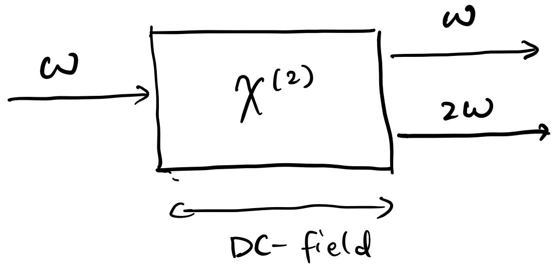
$$\tilde{E}(t) = E e^{-i\omega t} + E^{*} e^{i\omega t} = E e^{-i\omega t} + \underbrace{c.c.}_{\text{complex conjugate}}$$

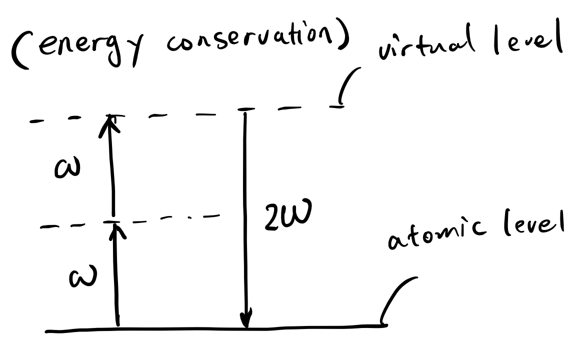
$$\tilde{P}^{(2)}(t) = \epsilon_0 \chi^{(2)} \tilde{E}^2(t)$$

$$= \underbrace{2\epsilon_0 \chi^{(2)} E E^{*}}_{\text{DC}} + \underbrace{(\epsilon_0 \chi^{(2)} E^2 e^{-i2\omega t}}_{\text{SHG}} + c.c.)$$

(Optical rectification, no EM radiation!)

Visualizations:





2 photons at ω are "destroyed"
one photon at 2ω is created

② Sum- and Difference-frequency Generation

Input fields:

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

$$\tilde{P}^{(2)}(t) = \epsilon_0 \chi^{(2)} \tilde{E}(t)^2$$

$$= \epsilon_0 \chi^{(2)} \Big[\underbrace{E_1^2 e^{-2i\omega_1 t}}_{SHG} + \underbrace{E_2^2 e^{-2i\omega_2 t}}_{SHG} + \underbrace{2E_1E_2 e^{-i(\omega_1+\omega_2)t}}_{SFG} + \underbrace{2E_1E_2^* e^{-i(\omega_1-\omega_2)t}}_{DFG} + c.c \Big]$$

$$+ 2\epsilon_0 \chi^{(2)} \underbrace{[E_1E_1^* + E_2 \cdot E_2^*]}_{OR.}$$

If we write $\tilde{P}^{(2)}(t) = \sum_n P(\omega_n) e^{-i\omega_n t}$

$$P(2\omega_1) = \epsilon_0 \chi^{(2)} E_1^2 \quad (SHG)$$

$$P(2\omega_2) = \epsilon_0 \chi^{(2)} E_2^2 \quad (SHG)$$

$$P(\omega_1+\omega_2) = 2\epsilon_0 \chi^{(2)} E_1 E_2 \quad (SFG)$$

$$P(\omega_1-\omega_2) = 2\epsilon_0 \chi^{(2)} E_1 E_2^* \quad (DFG)$$

Four nonlinear polarizations!

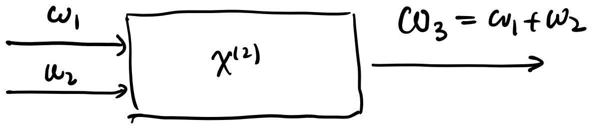
$$P(0) = 2\epsilon_0 \chi^{(2)} (E_1E_1^* + E_2E_2^*) \quad (OR)$$

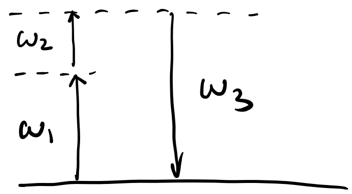
Comments:

Although in principle, four different non-zero freq. components are present in the nonlinear polarization, typically only one nonlinear process can occur efficiently. This is due to the constrains from phase matching (momentum conservation)

Sum-Frequency generation:

$$P(\omega_1+\omega_2) = 2\epsilon_0 \chi^{(2)} E_1 E_2$$

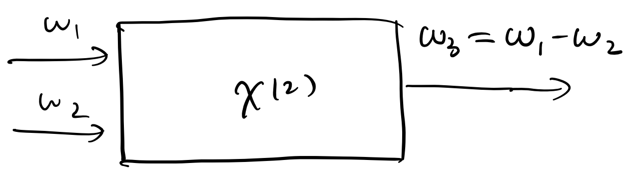


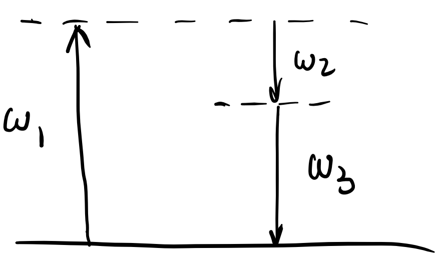


Application: generation of wavelength-tunable shorter wavelength light

Diffence-frequency generation and

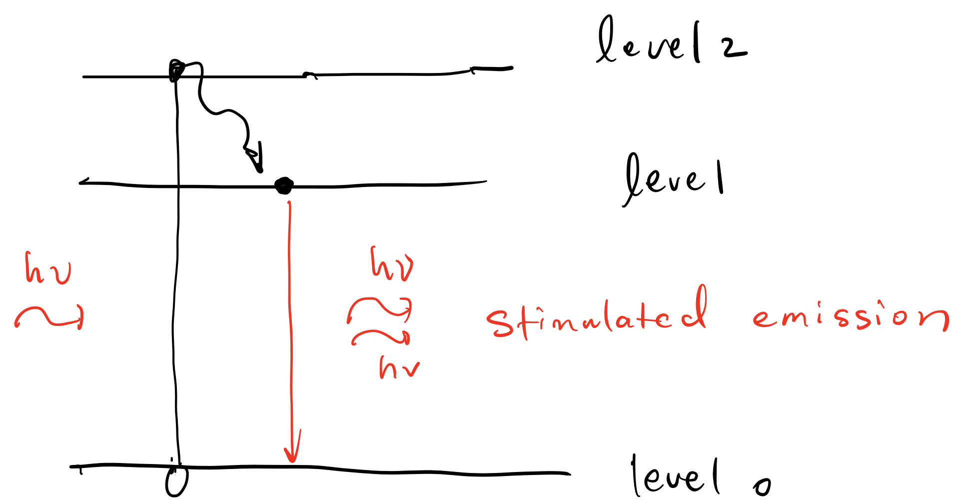
$$P(\omega_1-\omega_2) = 2\epsilon_0 \chi^{(2)} E_1 E_2^*$$



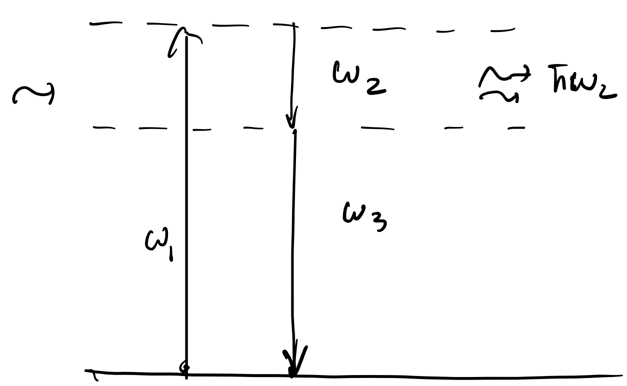


Comments: very similar to "three-level" laser system, so can work as an amplifier, a.k.a. <u>optical parametric amplification</u>. (OPA)

Three-level system in lasers



Similarly



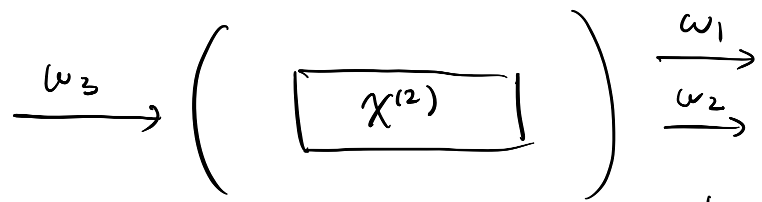
Physical meaning:

① the atom absorbs a photon of frequency ω_1 and jumps to the highest virtual level.

② the decay will be "stimulated" by the input at ω_2

③ Even without the input at ω_2, this down-conversion process can also happen, known as spontaneous parametric down conversion (SPDC), or parametric fluresceence.

Optical parametric oscillator (OPO)



tunable output by tuning the cavity!

③ The Pockels effect.

Input fields:

$$E = \underbrace{A_0}_{\text{DC-field}} + \underbrace{A_1 e^{-i\omega t}}_{\text{light-field}} + c.c.$$

$$P(t) = \epsilon_0 \left(\chi^{(1)} + 2\chi^{(2)} A_0\right) A_1 \cos(\omega t - kz) + \dots$$

Physical meaning:

index $n = \sqrt{1 + \chi^{(1)}}$ is changed to $\sqrt{1 + \chi^{(1)} + 2\chi^{(2)} A_0}$

Pockels effect only occurs in materials with $\chi^{(2)}$

Application: optical modulators.

4. Third-order nonlinear optical processes

3rd order nonlinear polarization:

$$\tilde{P}^{(3)}(t) = \epsilon_0 \chi^{(3)} \tilde{E}(t)^3$$

Consider the simplest input:

$$\tilde{E}(t) = A_0 + A_1 \cos(\omega t - kz)$$

using the identity. $\cos^3 \omega t = \frac{1}{4}\cos 3\omega t + \frac{3}{4}\cos\omega t$,

$$P(t) = \epsilon_0 \Big(\cdots + \underbrace{3\chi^{(3)} (A_0^2 + \tfrac{1}{4}A_1^2) A_1 \cos(\omega t - kz)}_{\text{index change}} + \underbrace{\tfrac{3}{2}\chi^{(3)} A_0 A_1^2 \cos(2\omega t - 2kz)}_{\text{DC-induced SHG}}$$

$$+ \underbrace{\tfrac{1}{4}\chi^{(3)} A_1^3 \cos(3\omega t - 3kz)}_{\text{THG.}} + \cdots$$

$$\Delta n \simeq \left(\frac{3\chi^{(3)}}{4c\epsilon_0 n^2} \right) I$$

Expand the first term

$$\underbrace{3\chi^{(3)} A_0^2 A_1 \cos(\omega t - kz)}_{\text{DC Kerr effect}} + \underbrace{\tfrac{3}{4}\chi^{(3)} A_1^3 \cos(\omega t - kz)}_{\substack{\text{AC Kerr effect} \\ \text{(intensity-dependent refractive index)}}}$$

5. NLO Zoology

	input	output	process
$\chi^{(2)}$	$\omega.\ \omega.$	$2\omega.$	SHG.
	$\omega,\ -\omega.$	$0.$	optical rectification
	$0.\ \omega.$	$\omega.$	Pockels effect.
	$\omega_1 \pm \omega_2$	$\omega_1 \pm \omega_2$	SFG, DFG OPA / OPO
$\chi^{(3)}$	$0\ \ 0\ \ \omega$	$\omega.$	Kerr-effect
	$0\ \ \omega.\ \omega$	2ω	DC-induced SHG
	$\omega.\ \omega.\ \omega.$	$3\omega.$	THG
	$\omega_1.\ \omega_2 \ldots \omega_n$	$n\omega_1$	High-harmonic generation