Nonlinear susceptibility

review linear theory:

$$\vec{D} = \vec{E} + 4\pi\vec{P}$$

$\vec{P}$ is polarization induced in medium by $\vec{E}$

if linear $\vec{P} = \chi^{(1)}\vec{E}$, $\chi^{(1)}$ = scalar if isotropic.

and

$$\vec{D} = (1 + 4\pi\chi^{(1)})\vec{E}$$
$$= \epsilon\vec{E}$$
$$\epsilon = n^2 = 1 + 4\pi\chi^{(1)}$$

In general, $\chi^{(1)}(\omega)$ with freq. dependence determined by resonance
- near resonance: complex Lorentzian.

In a linear medium, there is no coupling between freq. comps:

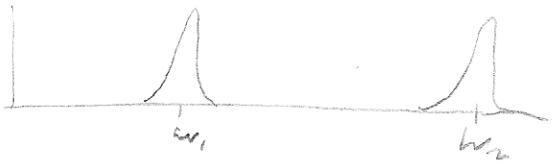
$$\vec{E}_{in} = \vec{E}_1 + \vec{E}_2 \qquad \text{where } \vec{E}_n = \underbrace{E_n(t)}_{\text{slowly varying}} e^{-i\omega n t}$$

or, $\mathcal{F}(\vec{E}_n(t)) = E_n(\omega)$

$$\rightarrow \vec{D}_{tot} = \left[1 + 4\pi\chi^{(1)}(\omega)\right]\left(\vec{E}_1(\omega) + \vec{E}_2(\omega)\right)$$

We're assuming $E_1(\omega)$ and $E_2(\omega)$ don't overlap:

"quasimonochromatic" E



$$\rightarrow \vec{D}_{tot} = \left(1 + 4\pi\chi^{(1)}(\omega_1)\right)\vec{E}_1(\omega_1) + \left(1 + 4\pi\chi^{(1)}(\omega_2)\right)E_2(\omega_2)$$
$$= \vec{D}_1 + \vec{D}_2$$

the waves propagate independently.

Nonlinear case

$$\vec{P}(t) = f(\vec{E}(t)) = \chi^{(1)}E + \chi^{(2)}E^2 + \chi^{(3)}E^3 + \cdots$$

series expansion: any $1/n!$ coeff are folded into $\chi^{(n)}$

how does wave eqn. change? (see section 2.1)

$$\nabla \times \vec{E} = -\frac{1}{c}\frac{\partial \vec{B}}{\partial t} \qquad \nabla \times \vec{H} = \frac{1}{c}\frac{\partial \vec{D}}{\partial t} + \frac{4\pi}{c}\frac{\partial \vec{J}}{\partial t}$$

$\vec{J} \to 0$

$$= -\frac{\mu}{c}\frac{\partial \vec{H}}{\partial t}$$

$$\nabla \times (\nabla \times \vec{E}) = -\frac{\mu}{c}\frac{\partial}{\partial t}(\nabla \times \vec{H}) = -\frac{\mu}{c}\frac{1}{c}\frac{\partial^2}{\partial t^2}\vec{D}$$

$$\nabla(\nabla \cdot \vec{E}) - \nabla^2\vec{E} = -\frac{\mu}{c^2}\frac{\partial^2 \vec{E}}{\partial t^2} - \frac{\mu}{c^2}4\pi\frac{\partial^2 \vec{P}}{\partial t^2}$$

$\nabla \cdot \vec{D} = 4\pi\rho \to 0$

linear case: $\vec{D} = \varepsilon\vec{E}$, if ε is spatially uniform,

$$\nabla \cdot (\varepsilon\vec{E}) = \varepsilon\,\nabla \cdot \vec{E} = 0$$

In NL case, $\nabla \cdot \vec{E} \neq 0$ in general, but is almost always small!

$$\to \nabla^2\vec{E} - \frac{\mu}{c^2}\frac{\partial^2 \vec{E}}{\partial t^2} = \frac{4\pi\mu}{c^2}\frac{\partial^2 \vec{P}}{\partial t^2}$$

split $\vec{P}$ into linear and nonlin parts $\vec{P} = \chi^{(1)}\vec{E} + \vec{P}^{NL}$

$$\to \nabla^2\vec{E} - \frac{\mu\varepsilon}{c^2}\frac{\partial^2 \vec{E}}{\partial t^2} = \frac{4\pi\mu}{c^2}\frac{\partial^2 \vec{P}^{NL}}{\partial t^2}$$

$\mu = 1$ (non magnetic)

$\mu\varepsilon = n^2$

$$\nabla^2 E - \frac{n^2}{c^2}\frac{\partial^2 \vec{E}}{\partial t^2} = \frac{4\pi}{c}\frac{\partial^2 \vec{P}^{NL}}{\partial t^2}$$

← source term

Nonlinear processes: (also see slides)

$\chi^{(n)}$ → different effects

usually treat these separately.

$\chi^{(2)}$: 2nd order

$P^{NL} = P^{(2)}(t) = \chi^{(2)} E^2$ (ignore vectors for now)

↳ squared, not $|\ |^2$!

representation of real fields:

in linear EM, often write

$\vec{E}(\vec{r},t) = \vec{E}_0 e^{i(\vec{k}\cdot\vec{r} - \omega t)}$ (complex)

and take $Re(\vec{E})$ at end.

in NL EM, must explicitly repr. fields as real.

$$E(t) = E_0 e^{-i\omega t} + E_0^* e^{i\omega t} \quad \text{(at } x=0\text{)}$$
$$= E_0 e^{-i\omega t} + c.c.$$

factors of $\frac{1}{2}$ matter:

we are not writing $E(t) = E_0 \cos(\omega t)$

$$= \tfrac{1}{2} E_0 \left(e^{i\omega t} + e^{-i\omega t}\right)$$

$$P^{(2)}(t) = \chi^{(2)} \left(E_0 e^{-i\omega t} + E_0^* e^{i\omega t}\right)^2$$

$$= 2\chi^{(2)} E_0 E_0^* + \chi^{(2)} E_0^2 e^{-i2\omega t} + \chi^{(2)} E_0^{*2} e^{i2\omega t}$$

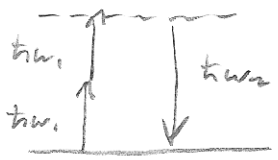
$2\chi^{(2)} E_0 E_0^*$: DC ($\omega = 0$), static field, "optical rectification"

$\chi^{(2)} E_0^2 e^{-i2\omega t} + \chi^{(2)} E_0^{*2} e^{i2\omega t}$: real source term at $\omega_2 = 2\omega_1$

Photon picture:

$\omega_2 = \omega_1 + \omega_1 \rightarrow \hbar\omega_2 = \hbar\omega_1 + \hbar\omega_1$

write schematically as:



SHG

→ dashed to repr. virtual level: no resonance.

opt. rect.

2 inputs:

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

$$P^{(2)} = \chi^{(2)} E^2 = \chi^{(2)}\left[E_1^2 e^{-i2\omega_1 t} + E_2^2 e^{-i2\omega_2 t} + 2E_1E_2 e^{-i(\omega_1+\omega_2)t} + 2E_1E_2^* e^{-i(\omega_1-\omega_2)t} + c.c.\right] + 2\chi^{(2)}\left[E_1E_1^* + E_2E_2^*\right]$$

now we have general outputs at diff't freq:

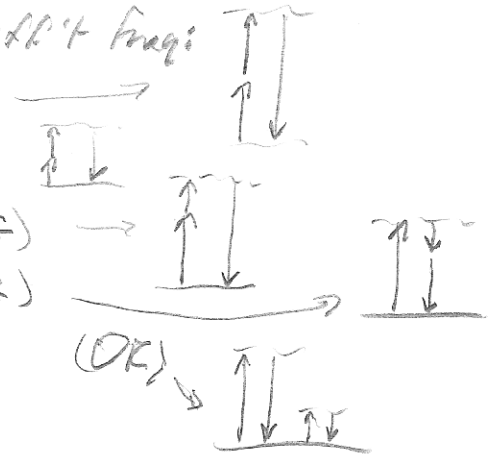
$P(2\omega_1) = \chi^{(2)} E_1^2$ } (SHG)

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

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

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

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



notice that E^* → how subtracted

Many processes: interesting, but complicated.

not all are optimized.

→ wave eqn for each frequency, coupled by source terms. (later)

Frequency generation devices:

- most lasers output 1 frequency, e.g. Nd:YAG $\lambda = 1.06\,\mu m$

SHG

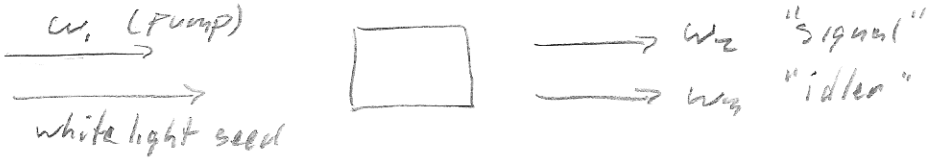
typ. 50% conv. eff.

can → near 100%

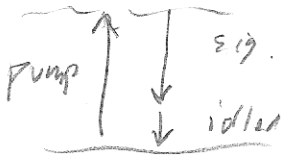
SFG: mix to get uv at $\omega_3 = \omega_2 + \omega_1$

at Rochester LLE ~ 75% at ω_3 from ω_1!

OPA: optical parametric amp.



w/ $\omega_1 = \omega_2 + \omega_3$



split photon.

Adjust crystal (phase matching) to optimize ω_2, ω_3

→ tunable output.

eg. start at $\lambda = 800\,nm$ (ti:sapphire)

→ 1-3 μm range. can double these → (visible)

DFG:

mix ω_2, ω_3 from OPA

→ 3-10 μm range