Nonlinear susceptibility

review linear theory:

$$\vec{D} = \vec{E} + 4\pi \vec{P} \quad (G) \qquad \boxed{\varepsilon_0 \vec{E} + \vec{P} \quad SI}$$

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

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

$$\boxed{\vec{P} = \varepsilon_0 \chi^{(1)} \vec{E} \quad SI}$$

and

$$\vec{D} = (1 + 4\pi \chi^{(1)}) \vec{E} \quad (G) \qquad \boxed{\vec{D} = \varepsilon_0 \vec{E} + \vec{P} = \varepsilon_0 (1 + \chi^{(1)}) \vec{E}}$$

$$= \varepsilon \vec{E}$$

$$\varepsilon = n^2 = 1 + 4\pi \chi^{(1)} \quad (G) \qquad \boxed{\varepsilon_{tot} = \varepsilon_0 \varepsilon \qquad \varepsilon = n^2 = 1 + \chi^{(1)}}$$

In general, $\chi^{(1)}(\omega)$ with freq. dependence determined by resonances

near resonance: complex Lorentzian

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

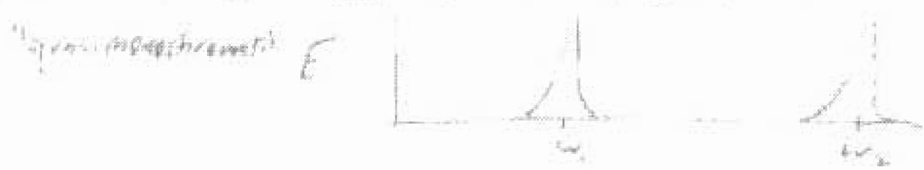
$$\vec{E}_{in} = \vec{E}_1 + \vec{E}_2 \qquad \text{where } \vec{E}_n = \underbrace{E_n(t)}_{\text{slowly varying}} e^{-i\omega_n t}$$

$$\text{or, } \mathcal{F}\{E_n(t)\} = E_n(\omega)$$

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

$$\boxed{\vec{D}_{tot} = \varepsilon_0 (1 + \chi^{(1)}(\omega))(\vec{E}_1(\omega) + \vec{E}_2(\omega))}$$

Let's assume $E_1(\omega)$ and $E_2(\omega)$ don't overlap:



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

$$= \vec{D}_1 + \vec{D}_2$$

the waves propagate independently.

Nonlinear case

$$\vec{P}(t) = f(\vec{E}(t)) = \epsilon_0(\chi^{(1)}E + \chi^{(2)}E^2 + \chi^{(3)}E^3 + \cdots)$$

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

how does wave eqn. change? [see section 2.1]

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

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: $\vec{D} = \epsilon\vec{E}$, if ϵ is spatially uniform,

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

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 $\vec{P} = \chi^{(1)}\vec{E} + \vec{P}^{NL}$

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

← source term

$\mu = 1$ (non magnetic)

$\mu\epsilon = n^2$

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

Nonlinear processes: (also see slides)

$\chi^{(n)} \to$ different effects

usually treat these separately.

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

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

$\hookrightarrow$ squared, not $| \, |^2$!

representation of real fields:

in linear E&M, often write

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

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

in NL E&M, must explicitly repr. fields as <u>real</u>

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

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

we are <u>not</u> writing $E(t) = E_0 \cos(\omega t)$
$$= \tfrac{1}{2} E_0 (e^{i\omega t} + e^{-i\omega t})$$

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

$$= \underbrace{2\chi^{(2)} E_0 E_0^*}_{\text{DC } (\omega=0)} + \underbrace{\chi^{(2)} E_0^2 e^{-i2\omega t} + \chi^{(2)} E_0^{*2} e^{i2\omega t}}_{\text{real source term at } \omega_2 = 2\omega_1}$$

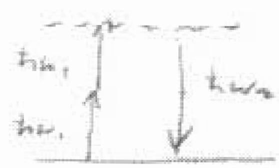
static field.

"optical rectification"

Photon picture:

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

write schematically as:



$\rightarrow$ dashed to repr. virtual level: no resonance

SHG

opt. rect

2 inputs:

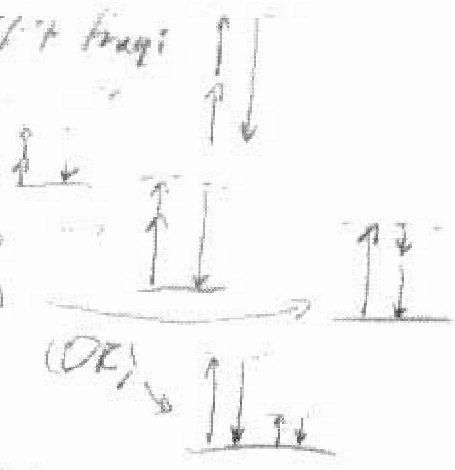
$$E(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + c.c.$$

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

$$\left. + 2E_1E_2^* e^{-i(\omega_1-\omega_2)t} + c.c.\right] + 2\chi^{(2)}[E_1E_1^* + E_2E$$

now we have general outputs at diff't freqs

$P(2\omega_1) = \chi^{(2)} E_1^2$
$P(2\omega_2) = \chi^{(2)} E_2^2$ } (SHG)
$P(\omega_1+\omega_2) = 2\chi^{(2)} E_1E_2$ (SFG)
$P(\omega_1-\omega_2) = 2\chi^{(2)} E_1E_2^*$ (DFG)
$P(0) = 2\chi^{(2)}(E_1E_1^* + E_2E_2^*)$ (OR)



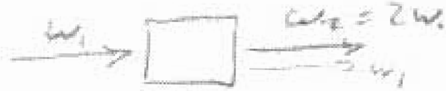
notice that $E^* \rightarrow \hbar\omega$ subtracted

Many processes: interesting, but complicated.
not all are optimized
$\rightarrow$ wave eqn for each frequency, coupled by source terms (later)

Frequency generation devices:

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

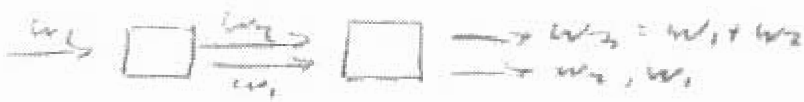
SHG



typ. 50% conv. eff.

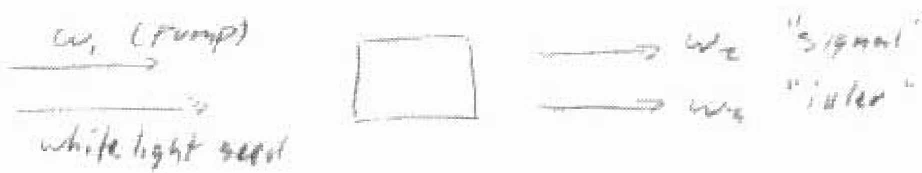
can → near 100%

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

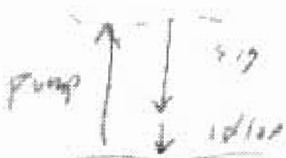


at Rochester LLE ~ 75% at ω_3 from ω_1!

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.

e.g. 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

3rd order effects, $\chi^{(3)}$

with ω_1 in, get:

THG

output at ω_1

→ change in refr. index

$$P^{(3)} \propto \chi^{(3)} E^3$$

$$P^{(3)} \propto \chi^{(3)} E E^* E$$

$$\sim \underbrace{\left(\chi^{(3)} I\right)}_{\sim n_2 I} E$$

write $n(I) = n_0 + n_2 I$

general case - "four-wave mixing"

example(s)

self-diffraction

$n(I(x)) \approx n_0 + n_2 I \cos(k_x x)$

NL index → phase grating

3rd photon scattering from grating

Sum-diff freq. mixing

$\omega_3 = 2\omega_2 - \omega_1$

self-focusing, self phase modulation, cross phase modulation

SF SPM XPM

and many more.

High-order: HHG