

Nonlinear processes:

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

usually treat these separately.

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

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

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

representation of real fields:

in linear EM, often write

$$\vec{E}(r, t) = \vec{E}_0 e^{i(E_0 r - \omega t)} \quad (\text{complex})$$

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

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

$$\begin{aligned} E(t) &= E_0 e^{-i\omega t} + E_0^* e^{i\omega t} \quad (\text{at } \alpha=0) \\ &= E_0 e^{-i\omega t} + \text{c.c.} \end{aligned}$$

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

$$\begin{aligned} \text{we are not writing } E(t) &= E_0 \cos(\omega t) \\ &= \frac{1}{2} E_0 (e^{i\omega t} + e^{-i\omega t}) \end{aligned}$$

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

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

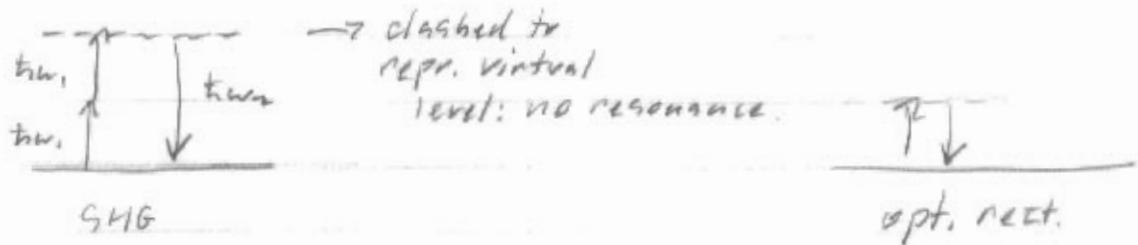
static field,

"optical rectification"

Photon picture:

$$\omega_2 = \omega_1 + \omega_1 \rightarrow h\omega_2 = h\omega_1 + h\omega_1$$

write schematically as:



2 inputs:

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

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

now we have general outputs at diff't freqs:

$$P(2\omega_1) = \chi^{(2)} E_1^{(2)} \quad \left. \right\} \text{(SHG)}$$

$$P(2\omega_2) = \chi^{(2)} E_2^{(2)} \quad \left. \right\}$$

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

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

$$P(0) = 2\chi^{(2)} (E_1 E_1^* + E_2 E_2^*) \quad \text{(DE)}$$

notice that $E^* \rightarrow \underline{h\nu}$ subtracted

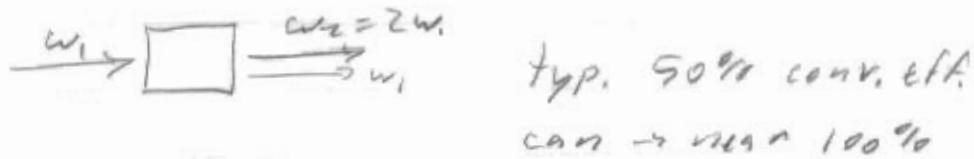
Many processes: interesting, but complicated.
not all are optimized.

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

Frequency generation devices:

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

SHG

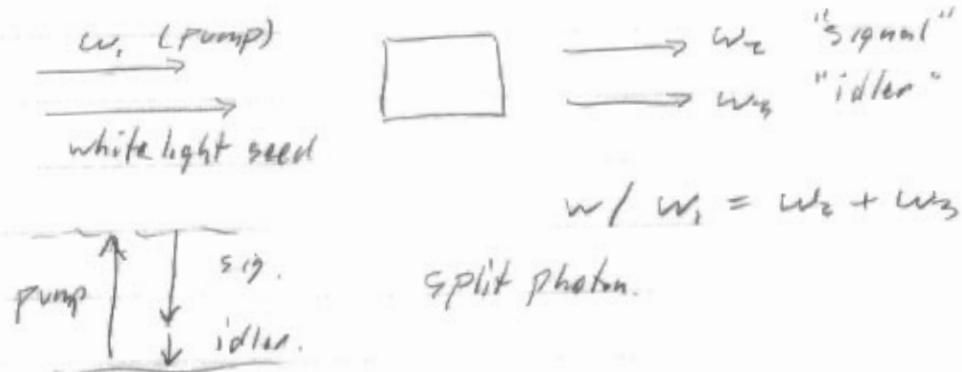


SFG : mix to get w_r at $w_3 = w_2 + w_1$



at Rochester LLE $\sim 75\%$ at w_3 , when w_1 !

OPA : optical parametric amp.



Adjust crystal (phase matching) to optimize w_2, w_3
 \rightarrow tunable output.

e.g. start at $\lambda = 800\text{ nm}$ (Li:apphire)

$\rightarrow 1 - 3\mu\text{m}$ range. can double these \rightarrow (visible)

PFG :

mix w_2, w_3 from OPA

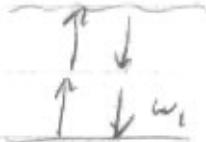
$\rightarrow 3 - 10\mu\text{m}$ range.

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

with ω_1 in, get:



THG



output at ω_3

$$P^{(2)} \propto \chi^{(3)} E^3$$

$$\begin{aligned} P^{(3)} &\propto \chi^{(3)} E E^* E \\ &\sim (\underbrace{\chi^{(3)} I}_{} E) E \\ &\sim n_2 I \end{aligned}$$

$$\text{write } n(I) = n_0 + n_2 I$$

general case: "Four-wave mixing"

examples



self-diffraction

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

NL index \rightarrow phase grating.

3rd photon scattering from grating.

Sum-diff freq. mixing

$$\frac{\omega_1}{\omega_2} \rightarrow \boxed{\quad} \rightarrow \omega_3 = 2\omega_2 - \omega_1$$

self-focusing, self-phase modulation, cross-phase modulation
SF SPM XPM

and many more.

High-order: HHG

Susceptibility (χ)

Linear case

induced polarization \propto applied field

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

↳ scalar if isotropic

tensor if anisotropic (birefringent)

$$\rightarrow \vec{P} \neq \vec{E}$$

$$\vec{D} = \epsilon_0 \vec{E} + \vec{P}$$

$$= \epsilon_0 (1 + \chi^{(1)}) \vec{E}$$

$$\underbrace{\vec{E}}_{\vec{E} = n^2} \quad n = \text{refractive index}$$

in a gas (low density) $\vec{P} = N_a \vec{p}$ w/ \vec{p} = dipole moment of molecule.

in condensed medium, apply local field corrections.

$\chi^{(1)}$ is dependent on frequency $\rightarrow \chi^{(1)}(\omega)$

- controlled by resonances.

Maxwell eqns. no free charges, non-magnetic

$$\nabla \cdot \vec{D} = \nabla \cdot \epsilon \vec{E} = 0 \quad \nabla \times \vec{E} = - \frac{\partial}{\partial t} \vec{B}$$

$$\nabla \cdot \vec{B} = 0 \quad \nabla \times \vec{B} = \mu_0 \frac{\partial}{\partial t} \vec{D} = \underbrace{\mu_0 \epsilon_0 \frac{\partial \vec{E}}{\partial t}}_{\propto} + \mu_0 \frac{\partial \vec{P}}{\partial t}$$

$$\nabla \times (\nabla \times \vec{E}) = - \frac{\partial}{\partial t} (\nabla \times \vec{B}) = - \frac{1}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} - \underbrace{\mu_0 \frac{\partial^2 \vec{P}}{\partial t^2}}_{\propto}$$

$$\nabla (\nabla \cdot \vec{E}) - \nabla^2 \vec{E}$$

o if $\epsilon \neq \epsilon_0 \propto$

$$\text{inhomogeneous wave eqn: } \nabla^2 \vec{E} - \frac{1}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} = \underbrace{\mu_0 \frac{\partial^2 \vec{P}}{\partial t^2}}_{\text{source term}}$$

when $\vec{P} = \epsilon_0 \chi^{(1)} \vec{E}$ (linear case)

$$\nabla^2 \vec{E} - \frac{1}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} = \frac{1}{c^2} \chi^{(1)} \frac{\partial^2 \vec{E}}{\partial t^2} \rightarrow \nabla^2 \vec{E} - \frac{n^2}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} = 0$$

if $\vec{E} = \vec{E}_1 + \vec{E}_2$ wave eqn separates into two independent eq
 \therefore no coupling.

Nonlinear case:

\vec{P} has a more complicated dependence on \vec{E}

often can expand in a Taylor series:

$$\vec{P} = \epsilon_0 \left(\chi^{(1)} \vec{E} + \chi^{(2)} \vec{E}^2 + \chi^{(3)} \vec{E}^3 + \dots \right)$$

any $1/n!$ factors are included in χ 's

ignoring vector qualities for now

separate into linear and NL parts

$$\vec{P} = \epsilon_0 \chi^{(1)} \vec{E} + \vec{P}^{NL}$$

$$\rightarrow \nabla^2 \vec{E} - \frac{n^2}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} = \underbrace{\mu_0 \frac{\partial^2}{\partial t^2} \vec{P}^{NL}}_{\text{source term.}}$$

Typically, we concentrate the analysis on one (or just a few) processes.

Notation: treat the full field as the sum of components that have a well-defined central freq. ω_n and direction \vec{k}_n

$$\vec{E}(\vec{r}, t) = \sum_{n>0} \vec{E}_n(\vec{r}, t)$$

$$\begin{aligned} \vec{E}_n(\vec{r}, t) &= \vec{E}_n^{(n,t)} \cos(\vec{k}_n \cdot \vec{r} - \omega_n t) \\ &= \vec{A}_n(\vec{r}, t) e^{i(\vec{k}_n \cdot \vec{r} - \omega_n t)} + \text{c.c.} \end{aligned}$$

with this convention $A_n = \frac{1}{2} \vec{E}_n$

$$\therefore \text{intensity} = I_n = \frac{1}{2} \epsilon_0 c n |\vec{E}_n|^2 = 2 \epsilon_0 c n |A_n|^2$$

$$\text{recall } \vec{S} = c \vec{E} \times \vec{H}$$

Now we can write the total field

$$\vec{E}(\vec{r}, t) = \sum_n \vec{A}_n(\vec{r}, t) e^{-i(E_n \cdot \vec{r} - w_n t)}$$

↳ positive and negative freq.

same for the polarization:

$$\vec{P}(\vec{r}, t) = \sum_n \vec{P}(w_n) e^{-i\omega_n t}$$

↳ does depend on \vec{r}, t
but no $e^{iE_n \cdot \vec{r}}$

Now extend definition of nonlinear polarization: $\chi^{(2)}$ example

$$P_i(w_n + w_m) = \sum_{\substack{i \\ \text{output freq.}}} \sum_{\substack{j k \\ \text{in } (nm)}} \chi_{ijk}^{(2)}(w_n + w_m; w_n, w_m) E_j(w_n) E_k(w_m)$$

$\sum_{\substack{j k \\ \text{in } (nm)}}$ → sum over combinations $w_n + w_m = \text{constant}$
 $\rightarrow i^{\text{th}}$ cartesian component (1, 2, 3) sum freqs: $w_3 = w_1 + w_2$
 $= w_1 + w_2$

$\chi_{ijk}^{(2)}$ = tensor $\chi_{121}^{(2)}$ → polariz. along X
input along Y, X

many components of $\chi_{ijk}^{(2)}$ are zero, or equal!

- look at crystal symmetry

when sum has identical terms → degeneracy factors.