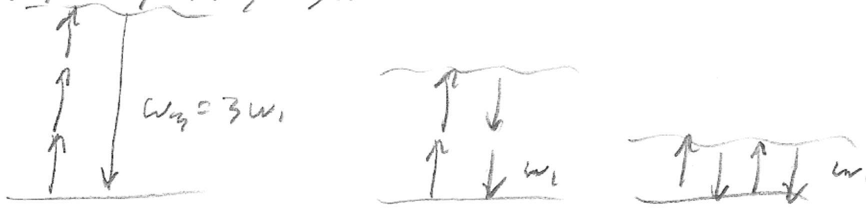
3rd order effects, $\chi^{(3)}$

with ω_1 in, get:



THG

output at ω_1

$\to$ 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: "fourwave mixing"

examples

self-diffraction

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

NL index $\to$ phase grating.

3rd photon scattering from grating.

sum-diff freq. mixing

self-focusing, self-phase modulation, cross-phase modulation

SF SPM XPM

and many more.

High-order: HHG

Semiclassical origins of $\chi^{(n)}$

- extend def of $\chi^{(n)}$ to include vector components of $\vec{E}$

$\chi^{(n)} \to$ tensor.

e.g. apply $\vec{E}$ along $\hat{x}$, can produce $\vec{P}$ along $\hat{y}$

here, define real field as

$$\vec{E}(\vec{r},t) = \vec{\mathcal{E}} \cos(\vec{k}\cdot\vec{r} - \omega t)$$

$$= \sum_n A(\omega_n) e^{i(\vec{k}_n\cdot\vec{r} - \omega_n t)}$$

$$\sum_n A(\omega_n) = \frac{1}{2}\mathcal{E}$$

n is +/-, with $A(\omega_n) = A^*(-\omega_n)$

(keep track of $\frac{1}{2}$ factor)

for $\chi^{(2)}$:

$$P_i(\underbrace{\omega_n + \omega_m}_{\text{output freq}}) = \sum_{jk}\sum_{(nm)} \chi^{(2)}_{ijk}(\omega_n+\omega_m;\omega_n,\omega_m) E_j(\omega_n) E_k(\omega_m)$$

$P_i \to$ i^{th} direction

$(nm) \to$ sum over comb. n,m s.t. $\omega_n+\omega_m$ = const.

$jk \to j,k = 1,2,3$

- not all terms are important
- symmetries $\to$ $\chi^{(2)}_{ijk}$'s that are equal or zero.
- summing over identical terms $\to$ "degeneracy factors"

Classical osc. model

$$F = m\ddot{x} = -eE(t) - m\omega_0^2 x - 2m\gamma\dot{x} - max^2$$

$\uparrow$ driving $\uparrow$ restoring SHO (harmonic) $\uparrow$ damping $\uparrow$ anharmonic

rearrange:

$$\ddot{x} + 2\gamma\dot{x} + \omega_0^2 x + ax^2 = -eE/m$$

understand this in terms of potentials:

$$U(x) = \underbrace{\tfrac{1}{2} m\omega_0^2 x^2}_{SHO} + \tfrac{1}{3} max^3$$

pot'l is now asymmetric
- still parabolic at low amplitude (if a is "small")

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

solve with a perturbation method:
basic idea is that the dominant response will be from the linear equation.
- use that soln in an equation for the NL correction.

Order parameter: λ

$$x = \lambda x^{(1)} + \lambda^2 x^{(2)} + \lambda^3 x^{(3)} + \ldots$$

and driving term is $-\frac{\lambda e E}{m}$

Now put these into equation $\rightarrow$ gather terms of same order

$\lambda \qquad \ddot{x}^{(1)} + 2\gamma \dot{x}^{(1)} + \omega_0^2 x^{(1)} = -eE/m$ linear eqn.

$\lambda^2 \qquad \ddot{x}^{(2)} + 2\gamma \dot{x}^{(2)} + \omega_0^2 x^{(2)} + a\left(x^{(1)}\right)^2 = 0$

etc. always keeping terms of equal power of λ

linear solution → standard model for refr. index:
- notice that even though we have sum of two inputs, since eqn is linear, soln is sum of two solns:

$$x^{(1)}(t) = x^{(1)}(\omega_1)e^{-i\omega_1 t} + x^{(1)}(\omega_2)e^{-i\omega_2 t} + \text{c.c.}$$

$$x^{(1)}(\omega_j) = -\frac{e}{m}E \frac{1}{\omega_0^2 - \omega_j^2 - 2i\omega_j\gamma} \equiv \frac{-eE}{mD(\omega_j)}$$

$D(\omega_j)$ = resonance denominator.

recall dipole is $p = qx \rightarrow -ex^{(1)}$
and polarization is

$$P = Np = -Ne\left(\frac{-eE(\omega_j)}{mD(\omega_j)}\right)$$

↳ # density

$$= \chi^{(1)}(\omega_j)E(\omega_j)$$

and, as usual, $\varepsilon = 1 + 4\pi\chi^{(1)}(\omega_j)$

Now $x^{(1)}(t)$ is treated as a known solution.
find $x^{(2)}(t)$:

$$\ddot{x}^{(2)} + 2\gamma \dot{x}^{(2)} + \omega_0^2 x^{(2)} = -a\left(\frac{-eE(\omega_1)}{mD(\omega_1)} + \frac{-eE(\omega_2)}{mD(\omega_2)} + \text{c.c.}\right)^2$$

linear eqn. (homog) = driving term.

As we've seen before, there are several terms that come out of the $(\ \ )^2$. Once the sq. term is expanded, the terms are additive.

$\therefore$ group terms according to osc. freq.

for example, look at $\omega_m = \omega_1 - \omega_2$:

RHS (source term) $= -\dfrac{a e^2}{m^2} \cdot \dfrac{2 E_1 E_2^*}{D(\omega_1) D(\omega_2)}$

solution is $x^{(2)}(\omega_1-\omega_2)\, e^{-i(\omega_1-\omega_2)t}$

- put this into LHS

$\rightarrow \left(-(\omega_1-\omega_2)^2 - i 2\gamma(\omega_1-\omega_2) + \omega_0^2\right) x^{(2)}(\omega_1-\omega_2)$

$= D(\omega_1-\omega_2)\, x^{(2)}(\omega_1-\omega_2)$

$$x^{(2)}(\omega_1-\omega_2) = \frac{-2a(e/m)^2 E_1 E_2^*}{D(\omega_1) D(-\omega_2) D(\omega_1-\omega_2)}$$

Final step: calc. $\chi^{(2)}$:

linear: $P^{(1)} = N(-e x^{(1)})$

2^{nd} order $P^{(2)} = N(-e x^{(2)}\underbrace{(\omega_1-\omega_2)}_{\text{or whichever combination.}})$

$= \chi^{(2)}(\omega_1-\omega_2;\ \omega_1, \omega_2)\, E(\omega_1) E^*(\omega_2)$

$$\chi^{(2)}(\quad) = \frac{2N(e^3/m^2)a}{D(\omega_1) D(-\omega_2) D(\omega_1-\omega_2)} = \frac{m a}{N^2 e^3} \chi^{(1)}(\omega_1-\omega_2) \chi^{(1)}(\omega_1) \chi^{(1)}(-\omega_2)$$

notes: • factor of 2 comes from permutations of distinct fields. ω_1, ω_2

• nonlinearity is enhanced by resonance.