

Classical driven oscillator model

- extension of model for linear dispersion
- electron bound to ion w/ spring $\vec{E}(t) \downarrow \uparrow$
 - linear + nonlinear restoring force
 - driven by $E(t)$ at given ω or sum of ω 's
 - damped (velocity-dependent)
- motion $x(t) \rightarrow$ induced dipole $P(t)$
macroscopic polarization: $P(t) = N_a P(t)$
 - $\chi^{(1)}$ from $P = \chi^{(1)} E$
 - $\chi^{(2)}$ from $\chi^{(2)} E^2$
 - etc

So we must solve for $x(t)$ with NL restoring force.

- no approx: numerical solns e.g. `NDSolve[]`
- perturbative soln

Later: QM approaches

- time dependent PT
- density matrix, Bloch eqn.

main differences have to do with resonances.

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:

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

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

ignoring vector
 qualities for now

Separate into linear and NL parts

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

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

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

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

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

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

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

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

Now we can write the total field

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

↳ positive and negative freq.

same for the polarization:

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

↳ does depend on \vec{r}, t

but no $e^{i\vec{k}_n \cdot \vec{r}}$

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

$$P_i(\omega_n + \omega_m) = \sum_{jk} \sum_{(nm)} \chi_{ijk}^{(2)}(\omega_n + \omega_m; \omega_n, \omega_m) E_j(\omega_n) E_k(\omega_m)$$

↳ output freq. $\omega_n + \omega_m = \text{constant}$
 ↳ $j, k = 1, 2, 3$
 ↳ i^{th} cartesian component (1, 2, 3)
 sum freq: $\omega_3 = \omega_1 + \omega_2 = \omega_2 + \omega_1$

$\chi_{ijk}^{(2)}$ = tensor $\chi_{121}^{(2)} \rightarrow$ 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 \rightarrow degeneracy factors.

Classical oscillator model: nonlinear response

individual dipole $\vec{p} = q\vec{r} = -eX\hat{x}$ in 1D
polarizability $\vec{P} = N_0\vec{p}$

eqn of motion - assume spring restoring force + x^3 term

$$F = m\ddot{x} = -eE(t) - m\omega_0^2 x - 2m\gamma\dot{x} - m\alpha x^3$$

driving term Hooke's damping anharmonic

$$\rightarrow \ddot{x} + 2\gamma\dot{x} + \omega_0^2 x + \alpha x^3 = -\frac{e}{m} E(t)$$

As potential:

$$U(x) = \frac{1}{2} m\omega_0^2 x^2 + \frac{1}{3} m\alpha x^3$$

asym pot'l



Input field:

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

Perturbation solution

- assume dominant response is from linear solution
- use linear solution to calculate NL correction
- similar to QM Pert Theory

Set an order parameter λ - just a device to keep track

$$X(t) = \lambda X^{(1)}(t) + \lambda^2 X^{(2)}(t) + \lambda^3 X^{(3)}(t) + \dots$$

driving term is $\propto \lambda$

\rightarrow eqn, gather terms of same order.

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

$$\lambda^2 \quad \ddot{X}^{(2)} + 2\gamma \dot{X}^{(2)} + \omega_0^2 X^{(2)} + a (X^{(1)})^2 = 0$$

etc. always keeping terms of equal power of λ

linear solution \rightarrow 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:

$$X^{(1)}(t) = X_0^{(1)}(\omega_1) e^{-i\omega_1 t} + X_0^{(1)}(\omega_2) e^{-i\omega_2 t} + \text{c.c.}$$

$$X_0^{(1)}(\omega_j) = -\frac{e}{m} E_j \frac{1}{\omega_0^2 - \omega_j^2 - 2i\omega_j\gamma} \equiv -\frac{eE_j}{m D(\omega_j)}$$

$D(\omega_j)$ = resonance denominator. ω_j can be + or -

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

and polarization is

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

\hookrightarrow # density

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

and, as usual, $\epsilon = 1 + 4\pi X^{(1)}(\omega_j)$ (S) $\epsilon = 1 + X^{(1)}(\omega_j)$ SI

Now $X^{(1)}(t)$ is treated as a known solution.

find $X^{(2)}(t)$:

$$\ddot{X}_0^{(2)} + 2\gamma \dot{X}_0^{(2)} + \omega_0^2 X_0^{(2)} = -a \left(\frac{-eE_1(\omega_1)}{m D(\omega_1)} + \frac{-eE_2(\omega_2)}{m D(\omega_2)} + \text{c.c.} \right)$$

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 $\omega_m = \omega_1 - \omega_2$:

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

solution is $X_0^{(2)}(\omega_1 - \omega_2) e^{-i(\omega_1 - \omega_2)t}$

- put this into LHS

$$\begin{aligned} \rightarrow (- (\omega_1 - \omega_2)^2 - i \gamma \delta(\omega_1 - \omega_2) + \omega_0^2) X_0^{(2)}(\omega_1 - \omega_2) \\ = D(\omega_1 - \omega_2) X_0^{(2)}(\omega_1 - \omega_2) \end{aligned}$$

$$X_0^{(2)}(\omega_1 - \omega_2) = \frac{-2a (e/m)^2 E_1 E_2^*}{D(\omega_1) D(-\omega_2) D(\omega_1 - \omega_2)}$$

enhancement: resonance on fundamental, output

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

linear: $P^{(1)} \equiv N(-e X^{(1)})$ both G, SI

2nd order: $P^{(2)} \equiv N(-e X^{(2)}(\omega_1 - \omega_2))$

or whichever combination

$$= X^{(1)}(\omega_1 - \omega_2; \omega_1, \omega_2) E(\omega_1) E^*(\omega_2)$$

$\times E_0$ for SI

$$X^{(2)}(\) = \frac{N (e^2/m^2) a}{(E_0) D(\omega_1) D(-\omega_2) D(\omega_1 - \omega_2)} = \frac{m a}{2^2 3} \frac{X^{(1)}(\omega_1 - \omega_2) X^{(1)}(\omega_1)}{N e (E_0) X^{(1)}(\omega_1)}$$

notes: • factor of 2 comes from permutations of distinct fields ω_1, ω_2 (this is for SI only)

• non-linearity is enhanced by resonance.