

Review: non isotropic, linear medium (birefringent)
 - different refractive index for different directions.

isotropic: $\vec{D} = \epsilon \vec{E} = (1 + 4\pi\chi^{(1)}) \vec{E}$ $\vec{E} \parallel \vec{D}$

non-isotropic

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

$$= \begin{pmatrix} \epsilon_{xx} & \epsilon_{xy} & \epsilon_{xz} \\ \epsilon_{yx} & \epsilon_{yy} & \epsilon_{yz} \\ \epsilon_{zx} & \epsilon_{zy} & \epsilon_{zz} \end{pmatrix} \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix}$$

Now \vec{D} isn't parallel to \vec{E}
 or, since $\vec{D} = \vec{E} + 4\pi\vec{P}$
 $= \vec{E} + 4\pi\vec{\chi} \cdot \vec{E}$

$$\vec{D} = \epsilon_0 \vec{E} + \vec{P} \quad \text{SI}$$

$$= \epsilon_0 (\vec{E} + \vec{\chi} \cdot \vec{E}) \quad \text{CGS}$$

Apply \vec{E} , response is along \vec{P} ,
 could write as:

$$P_i(\omega) = \sum_j \chi_{ij}(\omega) E_j(\omega)$$

It is possible to find a basis (coord. system) in which
 $\vec{\epsilon}$ or $\vec{\chi}^{(1)}$ is diagonal. These are the crystal axes.
 in this basis:

$$\vec{D} = \begin{pmatrix} \epsilon_x & & \\ & \epsilon_y & \\ & & \epsilon_z \end{pmatrix} \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix}$$

note: crystal structure
 doesn't have to be anythg
 special to diagonalize

$\epsilon_x = \epsilon_y = \epsilon_z$ isotropic, otherwise, biaxial
 $\epsilon_x = \epsilon_y \neq \epsilon_z$ uniaxial

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: $P(t) = N_0 p(t)$

$\chi^{(1)}$ 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 solns

Later: QM approaches

- time dependent PT
- density matrix, Bloch eqn.

main differences have to do with resonances.

Classical osc. model: dipole $\vec{p} = q\vec{r} \rightarrow P_x = -eX \rightarrow P_x = N_A p_x$

$$F = m\ddot{x} = \underbrace{-eE(t)}_{\text{driving}} - \underbrace{m\omega_0^2 X}_{\substack{\text{restoring} \\ \text{SHO} \\ \text{(harmonic)}}} - \underbrace{2m\delta\dot{X}}_{\text{damping}} - \underbrace{m\alpha X^2}_{\text{anharmonic}}$$

rearrange:

$$\ddot{X} + 2\delta\dot{X} + \omega_0^2 X + \alpha X^2 = -eE/m$$

understand this in terms of potentials:

$$U(x) = \underbrace{\frac{1}{2} m\omega_0^2 X^2}_{\text{SHO}} + \frac{1}{3} m\alpha X^3$$



pot'l is now asymmetric

- still parabolic at low amplitude (if α is "small")

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

solve with a perturbation method:

basic idea is that the dominant response will be from the linear equation.

- use that to get in an equation for the NL corrections.

Order parameter: λ

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

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

Now put these into equation \rightarrow 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 retro. index:

- notice that even though we have sum of two inputs, since eqn is linear, soln is sum of two solns:

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

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

$D(\omega_j)$ = 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) $\epsilon = 1 + X^{(1)}(\omega)$ (S)

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

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

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

linear eqn (homog) = driving term.

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

\therefore group terms according to osc. freq.

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

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

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

- put that into LHS

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

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

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

linear: $P^{(1)} = N(-e x^{(1)})$ both G, SI

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

or whichever combination.

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

$\times E_0$ for SI

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

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

• non-linearity is enhanced by resonance.