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

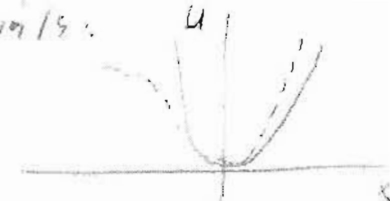
$$F = m\ddot{x} = \underbrace{-eE(t)}_{\text{driving}} \underbrace{- m\omega_0^2 x}_{\substack{\text{restoring} \\ \text{SHO} \\ \text{(harmonic)}}} \underbrace{- 2m\gamma\dot{x}}_{\text{damping}} \underbrace{- m a x^2}_{\text{anharmonic}}$$

rearrange:

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

understand this in terms of potentials:

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



pot'l is now asymmetric

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

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

solve with a perturbative 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

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

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

Now put these into equation → 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:

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

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

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

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

$$P = \underbrace{N}_{\text{\# density}} p = -Ne\left(-\frac{eE(\omega_j)}{mD(\omega_j)}\right)$$

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

and, as usual, $\epsilon = 1 + 4\pi\chi^{(1)}(\omega_j)$ (G) $\boxed{\epsilon = 1 + \chi^{(1)}(\omega_j) \text{ SI}}$

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

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

linear eqn (homog) = driving term.

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

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

for example, look at $\omega_{\pm} = \omega_1 - \omega_2$:

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

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

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

$\times E_0$ for SI

$$\chi^{(2)}(\quad) = \frac{2N(e^3/m^2)\,a}{(\varepsilon_0)\, D(\omega_1) D(-\omega_2) D(\omega_1 - \omega_2)} = \frac{m a}{N^2 e^3 (\varepsilon_0)} \chi^{(1)}(\omega_1 - \omega_2)\, \chi^{(1)}(\omega \ldots)\, \chi^{(1)} \ldots$$

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

• nonlinearity is enhanced by resonance.

Miller's rule

notice that

$$\chi^{(2)}(\omega_1-\omega_2;\omega_1,\omega_2) \propto \chi^{(1)}(\omega_1)\,\chi^{(1)}(-\omega_2)\,\chi^{(1)}(\omega_1-\omega_2)$$

ratio is $\dfrac{ma}{N^2 e^3 \varepsilon_0}$

Empirically this is generally true.

$\therefore$ measure linear index $\rightarrow$ NL coeff.

can also estimate size of $\chi^{(2)}$

N = num. density $\sim 1/d^3$ $\quad$ d = atomic spacing

$m = m_e$, e = charge

a = coupling constant

Estimate from potentials:

when amplitude $x \sim d$ $\quad$ linear, NL terms are ~equal

$$\tfrac{1}{2} m \omega_0^2 x^2 \approx \tfrac{1}{3} m a x^3$$

$$\rightarrow a \sim \frac{3}{2}\frac{\omega_0^2}{d}$$ $\quad$ ω_0 = resonance freq. (absorption begins)

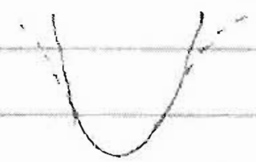
Same treatment can be used to estimate $\chi^{(3)}$

Centro-symmetric media

- see details in book

restoring force model: $\quad -m\omega_0^2\vec{r} + mb(\vec{r}\cdot\vec{r})\vec{r}$

- at large $|r|$, less binding
- force is always along $\hat{r}$



Perform pert. expansion.

- note $X^{(2)}$ equation has no driving term

$\therefore X^{(2)} = 0$, any initial value of $X^{(2)}(t=0)$ damps

$$\ddot{X}^{(3)} + 2\gamma\dot{X}^{(3)} + \omega_0^2 X^{(3)} = b\left(X^{(1)}\right)^3$$

↳ linear solution here.

$$\rightarrow \vec{r}^{(3)}(\omega_q) = -\sum_{(mnp)} \frac{be^3}{m^3} \frac{\left(\vec{E}(\omega_m)\cdot\vec{E}(\omega_n)\right)\vec{E}(\omega_p)}{D(\omega_q)D(\omega_m)D(\omega_n)D(\omega_p)}$$

solution follows from parallel to force eqn.

resonance structure as usual

Vector / Tensor representation of $P^{(2)}$

index form:

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

n, m index input freq

i, j, k index Cartesian components.

ex.

$$P_x^{(2)}(\omega_3=\omega_1+\omega_2) = (E_{x1}, E_{y1}, E_{z1}) \underbrace{\begin{pmatrix} \chi_{xxx} & \chi_{xxy} & \chi_{xxz} \\ \chi_{xyx} & \chi_{xyy} & \chi_{xyz} \\ \chi_{xzx} & \chi_{xzy} & \chi_{xzz} \end{pmatrix}}_{@\,(\omega_1+\omega_2;\,\omega_1,\omega_2)} \begin{pmatrix} E_{x2} \\ E_{y2} \\ E_{z2} \end{pmatrix}$$

$$+ (E_{x2}\ E_{y2}\ E_{z2}) \underbrace{\begin{pmatrix} & \overleftrightarrow{\chi} & \end{pmatrix}}_{@\,(\omega_2+\omega_1;\,\omega_2,\omega_1)} \begin{pmatrix} \\ \\ \end{pmatrix}$$

swap 1 ⟷ 2

$$= \sum_{(nm)} \vec{E}_n \cdot \overleftrightarrow{\chi}(\omega_n+\omega_m;\omega_n,\omega_m)\cdot \vec{E}_m$$

Also need $-\omega_3 = -\omega_1 + -\omega_2$ terms

and P_y, P_z

this is only for $\omega_3 = \omega_1+\omega_2$ process!

Take advantage of symmetries → many $\chi^{(n)}$ component are zero or equal.