

Miller's rule

notice that

$$\chi^{(2)}(w_1 - w_2; w_1, w_2) \propto \chi^{(1)}(w_1) \chi^{(1)}(-w_2) \chi^{(4)}(w_1 - w_2)$$

ratio is $\frac{ma}{N^2 e^3 \epsilon_0}$

Empirically this is generally true,

∴ measure linear index \rightarrow NL coeff.

can also estimate size of $\chi^{(1)}$

$$N = \text{nvm. density} \sim 1/d^3 \quad d = \text{atomic spacing}$$

$$m = m_e, e = \text{charge}$$

$$a = \text{coupling constant}$$

Estimate from potentials:

when amplitude $X \sim d$ linear, NL terms are negl

$$\frac{1}{2} m w_0^2 X^2 \approx \frac{1}{2} m a X^3$$

$$\rightarrow a \sim \frac{3}{2} \frac{w_0^2}{d} \quad w_0 = \text{resonance freq.}$$

(absorption begins)

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

Centro-symmetric media

- see details in book

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

- at large $|\vec{r}|$, less binding

- force is always along \vec{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

$$\ddot{\vec{X}}^{(1)} + 2\gamma \dot{\vec{X}}^{(1)} + \omega_0^2 \vec{X}^{(1)} = \mathbf{b}(\vec{X}^{(1)})$$

\hookrightarrow linear solution here.

$$\Rightarrow \vec{r}^{(1)}(\omega_q) = - \sum_{(mnp)} \frac{b e^i}{m^q} \frac{(\vec{E}(\omega_m) \cdot \vec{E}(\omega_n)) \vec{E}(\omega_p)}{D(\omega_q) D(\omega_m) D(\omega_n) D(\omega_p)}$$

solution follows from parallel to force eqn.

resonance structure as usual

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

Index form:

$$P_i^{(2)}(w_n + w_m) = \sum_{j,k} \sum_{(nm)} \chi_{ijk}^{(2)}(w_n + w_m; w_n, w_m) E_j(w_n) E_k(w_m)$$

n, m index input freq

i, j, k index Cartesian components.

$$\text{ex: } P_x^{(2)}(w_3 = w_1 + w_2) = (E_{x1}, E_{y1}, E_{z1}) \begin{pmatrix} \chi_{xxx} & \chi_{xxy} & \chi_{xxz} \\ \chi_{xyx} & \chi_{xyy} & \chi_{xyz} \\ \chi_{zxz} & \chi_{xzy} & \chi_{xzz} \end{pmatrix} \begin{pmatrix} E_{x2} \\ E_{y2} \\ E_{z2} \end{pmatrix}$$

$$+ (E_{x2}, E_{y2}, E_{z2}) \begin{pmatrix} \chi_{xxy} \\ \chi_{xyx} \\ \chi_{zyz} \end{pmatrix} \quad \text{group } i \neq j, l \quad \text{at } (w_n + w_l; w_n, w_l)$$

$$= \sum_{(nm)} \vec{E}_n \cdot \vec{\chi}(w_n + w_m; w_n, w_m) \cdot \vec{E}_m$$

Also need $-w_3 = -w_1 + -w_2$ terms

and P_y, P_z

this is only for $w_3 = w_1 + w_2$ process!

Take advantage of symmetries \rightarrow many $\chi^{(2)}$ component are zero or equal

Symmetry properties of $\chi^{(2)}$

- huge number of tensors, tensor components to specify in general,

$$\chi_{ijk}^{(2)}(w_1, w_2, w_3) + \chi_{ijk}^{(2)}(w_2, w_3, w_1)$$

and all permutations of w_i , and their negatives.

- symmetries reduce from 3724 ($\chi^{(2)}$) to 10 or less.

- * reality of fields:

$P(\vec{r}, t)$ is measurable, and real

$$\therefore P_a(-w_n - w_m) = P_a(w_n + w_m)^*$$

E_g are also real.

$$E_g(w_n) = E_g(w_m)^*$$

$$P_a(w_n + w_m)^* = \sum_{jk} \sum_{inms} \underbrace{\chi_{ijk}^{(2)}(w_n + w_m; w_n, w_m)}_{\text{real}} \bar{E}_j(w_n) \bar{E}_k(w_m)^*$$

$$= P_a(-w_n - w_m) = \sum_{jk} \sum_{inms} \chi_{ijk}^{(2)}(-w_n - w_m; -w_n, -w_m) E_j(-w_n) \bar{E}_k(-w_m)$$

χ can be complex, but this says w_n appears in χ_{ikm} as $i \neq n$

- * Intrinsic permutation symmetry: i, j, k are dummy indices

$$\begin{aligned} & \chi_{ijk}^{(2)}(w_n + w_m; w_n, w_m) E_j(w_n) E_k(w_m) \\ &= \chi_{ikj}^{(2)}(w_n + w_m; w_m, w_n) E_k(w_n) E_j(w_m) \end{aligned}$$

- swap $m \leftrightarrow n$ and $j \leftrightarrow k$ at same time.

- this applies only to the last 2 indices

- in the end, we sum up terms \rightarrow factor of 2 on this value of $\chi^{(2)}$

vector form $\vec{E}(w_n) \cdot \vec{\chi}_i \cdot \vec{E}(w_n) = \vec{E}(w_n) \cdot \vec{\chi}_i^T \cdot \vec{E}(w_m)$

- Full permutation symmetry: lossless medium only
 - all compn. of $\chi^{(2)}$ \rightarrow real
 - requires $|w_3 - w_0| \gg \gamma$ many linewidths away from any reson.
 - full planar symmetry: interchange any w 's along w/ corres. i, j, k
- $$\chi_{ijk}^{(2)}(w_3 = w_1 + w_2) = \chi_{jki}^{(2)}(w_1 = w_2 + w_3) \quad \begin{matrix} \text{permute} \\ \text{cyclic} \end{matrix}$$
- (comes from energy argument)
- $$= \chi_{jki}^{(2)}(w_1 = -w_0 + w_3)^* \quad P \in \text{real}$$
- $$= \chi_{jki}^{(2)}(w_1 = -w_2 + w_0) \quad \chi \text{ real}$$
- likewise,
- $$= \chi_{kij}^{(2)}(w_2 = w_3 - w_1) \quad \begin{matrix} 2 \uparrow \\ 1 \downarrow \end{matrix} \quad \begin{matrix} 3 \downarrow \\ 1 \uparrow \end{matrix} = \begin{matrix} 3 \uparrow \\ 2 \downarrow \end{matrix} \quad \begin{matrix} 1 \downarrow \\ 2 \uparrow \end{matrix}$$
- Kleinman's symmetry: dispersionless χ
- requires $w_a \ll w_0$ (could be other way too)
- \rightarrow instantaneous response: $P(\epsilon) = \chi^{(2)} E(\epsilon)^2$
- recall dispersion \equiv non-constant freq. response $H(\omega)$
- \rightarrow impulse response $+ \delta(t)$

Now we can permute ijk w/o permuting w 's

Next, all the matrices are symmetric

- see slide,

Contracted notation

If Kleinman's symmetry is valid, each of the three matrices are symmetric:

$$\begin{bmatrix} \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot \end{bmatrix}, \begin{bmatrix} \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot \end{bmatrix}, \begin{bmatrix} \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot \end{bmatrix} = 18 \text{ elements.}$$

contract second pair of indices to 1:

$$\begin{array}{ccccccccc} jk & 1, & 22 & 33 & 23,32 & 41,13 & 12,21 \\ l & \downarrow & 1 & 2 & 3 & 4 & 5 & 6 \end{array}$$

$$\rightarrow d_{11} = \begin{bmatrix} \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot \end{bmatrix} \text{ now just one matrix.}$$

not all are independent: can permute indices \rightarrow max 10 independent
- see slide

Crystal symmetry further reduces the # of independent and non-zero matrix elements.

32 possible crystal point groups

21 are non-centrosymmetric, \rightarrow no second-order
I² polarized.

w/ inversion symmetry, $\chi^{(2)} \rightarrow 0$

SIG example

$$P(t) = \chi^{(2)} E(t)^2 \quad E(t) = E_0 \cos \omega t$$

if $E(t) \rightarrow -E(t)$ (by inverting coordinate system)

$P(t) \rightarrow -P(t)$ with inversion symmetry.

$$\text{but } P(t) = \chi^{(2)} (-E(t))^2 = ? - P(t)$$

$$\therefore \chi^{(2)} = 0$$

similar arguments for crystal symmetries

Using the d-matrix to calculate d_{eff}

Phase matching usually dictates the allowed directions.

example: "Type I" phase matching has input as o-wave (n is independent of angle), output as e-wave $n = f(\theta)$,

Optic axis = z-axis

o-wave has no z-component

$$\vec{E}_{in} = \{E_x, E_y, 0\}$$

$$\vec{E}_{in} \cdot \vec{k} = 0 \quad \rightarrow \text{components } E_x, E_y \text{ for input}$$

e-wave is $\perp \vec{E}_{in}$ and $\perp \vec{k}$ \rightarrow

$$\begin{aligned} \vec{E}_{out} \cdot \vec{E}_{in} &= 0 \\ \vec{E}_{out} \cdot \vec{k} &= 0 \end{aligned} \quad \left. \begin{array}{c} \vec{E}_{out} \\ \vec{E}_{out} \end{array} \right\}$$

induced $\vec{P}_{in} = \left[\begin{array}{c} \cdot \cdot d \\ \vdots \end{array} \right] \left[\begin{array}{c} E_{1x} E_{2x} \\ E_{1y} E_{2y} \\ E_{1z} E_{2z} \\ E_{1z} E_{2y} + E_{1y} E_{2z} \\ \vdots \end{array} \right]$

use unit vectors for E_{in} 's

this \vec{P}_{in} has components in all directions

component along \vec{E}_{out} is what $\rightarrow d_{eff}$

$$d_{eff} = \vec{P}_{in} \cdot \vec{E}_{out}$$