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

- huge number of tensors, tensor components to specify

in general,

$$\chi^{(2)}_{ijk}(\omega_1, \omega_2, \omega_3) \neq \chi^{(2)}_{ijk}(\omega_2, \omega_3, \omega_1)$$

and all permutations of ω_i, and their negatives.

- symmetries reduce from 324 ($\chi^{(2)}$!) to 10 or less.

• reality of fields:

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

$$\therefore P_i(-\omega_n - \omega_m) = P_i(\omega_n + \omega_m)^*$$

E's are also real:

$$E_j(-\omega_n) = E_j(\omega_n)^*$$

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

$$= P_i(-\omega_n-\omega_m) = \sum_{jk}\sum_{(nm)} \overbrace{\chi^{(2)}_{ijk}(-\omega_n-\omega_m; -\omega_n, -\omega_m)} E_j(-\omega_n) E_k(-\omega_m)$$

χ can be complex, but this says ω_n appears in $\chi(\omega_n)$ as $i\omega_n$

• Intrinsic permutation symm. i, j, k are dummy indices

$$\chi^{(2)}_{ijk}(\omega_n+\omega_m; \omega_n, \omega_m) E_j(\omega_m) E_k(\omega_n)$$

$$= \chi^{(2)}_{ikj}(\omega_n+\omega_m; \omega_m, \omega_n) E_k(\omega_n) E_j(\omega_m)$$

- 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 $\quad \vec{E}(\omega_m) \cdot \overleftrightarrow{\chi}_i \cdot \vec{E}(\omega_n) = \vec{E}(\omega_n) \cdot \overleftrightarrow{\chi}_i^{T} \cdot \vec{E}(\omega_m)$

- Full permutation symmetry: ~~lossless~~ medium only
  - all compon. of $\chi^{(n)} \to$ real

    requires $|\omega_n - \omega_0| >> \gamma$ many linewidths away from any reson.

  - full perm. symm: interchange any ω's along w/ corresp. i, j, k

$$\chi^{(2)}_{ijk}(\omega_3 = \omega_1 + \omega_2) = \chi^{(2)}_{jki}(-\omega_1 = \omega_2 - \omega_3) \quad \text{permute. cyclic}$$

(comes from energy argument) 1.5.4

$$= \chi^{(2)}_{jki}(\omega_1 = -\omega_2 + \omega_3)^* \quad P(t) \text{ real}$$

$$= \chi^{(2)}_{jki}(\omega_1 = -\omega_2 + \omega_3) \quad \chi \text{ real}$$

likewise; $= \chi^{(2)}_{kij}(\omega_2 = \omega_3 - \omega_1)$ $2\uparrow\ 1\uparrow\ \downarrow 3 = 3\uparrow\ \downarrow 1\ \downarrow 2 = $

- Kleinman's symmetry: dispersionless χ

  requires $\omega_n << \omega_0$ (could be other way too)

  $\to$ instantaneous response: $P(t) = \chi^{(2)} E(t)^2$

  recall dispersion = non-constant freq. response $H(\omega)$

  $\to$ impulse response $\neq \delta(t)$

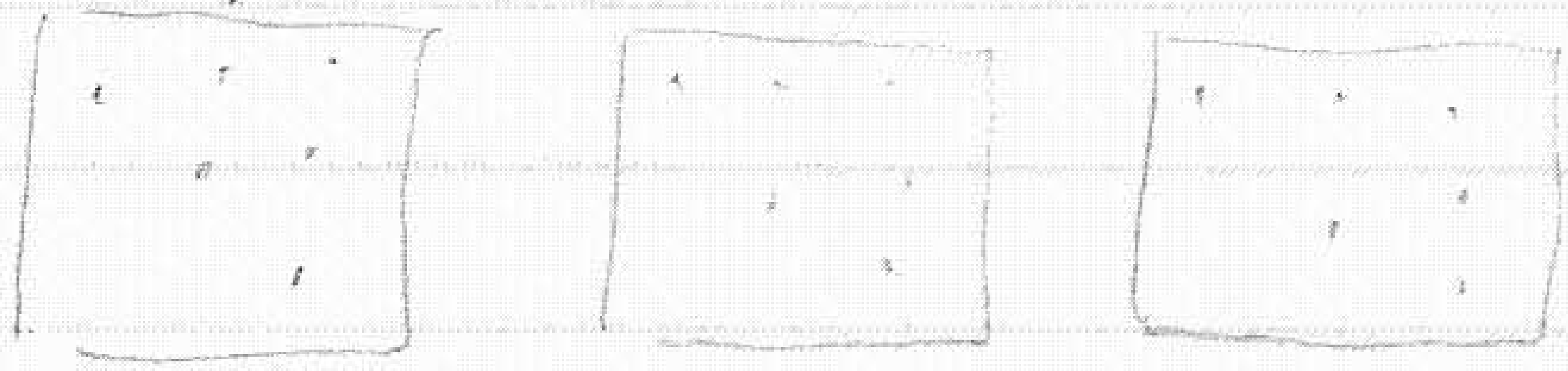
Now we can permute ijk w/o permuting ω's

Here, all the matrices are symmetric

- see slide.

Contracted notation

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

 = 18 elements.

contract second pair of indices to l:

jk	11	22	33	23,32	31,13	12,21
l	1	2	3	4	5	6

$\rightarrow d_{il} =$  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 response.

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

SHG example

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

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

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

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

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

similar arguments for crystal symmetries

Effective d:

actual orientation of fields is restricted by phase matching:

for $\omega_3 = \omega_1 + \omega_2$

$k_3 = k_1 + k_2$ or $\Delta k = k_1 + k_2 - k_3 = 0$

within this restriction → cone of angles.

- value of d_{eff} determines best choice.
- we'll return to this when we treat phase matching.