Nonlinear Optics
Summary of Lecture 2

3. 10. 2018

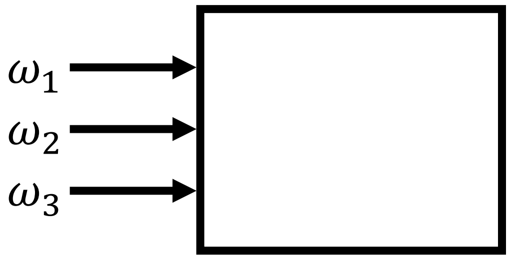
Formal Definition of the Nonlinear Susceptibility

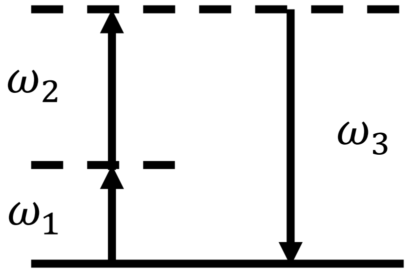
- $\tilde{\boldsymbol{E}}(\boldsymbol{r},t) = \sum_n \boldsymbol{E}(\omega_n)e^{-i\omega_n t} = \sum_n A(\omega_n)e^{i(\boldsymbol{k_n r}-\omega_n t)}$
- $\tilde{\boldsymbol{P}}(\boldsymbol{r},t) = \sum_n \boldsymbol{P}(\omega_n)e^{-i\omega_n t}$
- $\boldsymbol{E}^*(\omega_n) = \boldsymbol{E}(-\omega_n)$
  $\boldsymbol{P}^*(\omega_n) = \boldsymbol{P}(-\omega_n)$

Response for $\boldsymbol{\omega_m}+\boldsymbol{\omega_n}$:

- $\tilde{\boldsymbol{P}}(\boldsymbol{r},t) = \sum_{(n,m)} \boldsymbol{P}(\omega_n+\omega_m)e^{-i(\omega_n+\omega_m)t}$
- $\chi^{(2)}$ defines the amplitude of P
- $P_i(\omega_n+\omega_m) = \varepsilon_0 \sum_{jk} \sum_{(n,m)} \chi^{(2)}_{ijk}(\omega_n+\omega_m;\omega_n,\omega_m)E_j(\omega_n)E_k(\omega_m)$

Properties of $\chi^{(2)}$





$$\omega_3 = \omega_1 + \omega_2$$
$$\omega_3 = \omega_2 + \omega_1$$

$$\omega_1 = \omega_3 - \omega_2$$
$$\omega_1 = -\omega_2 + \omega_3$$
$$\omega_2 = \omega_3 - \omega_1$$
$$\omega_2 = -\omega_1 + \omega_3$$

$$-\omega_3 = -\omega_1 - \omega_2$$
$$-\omega_3 = -\omega_2 - \omega_1$$
$$-\omega_1 = -\omega_3 + \omega_2$$
$$-\omega_1 = \omega_2 - \omega_3$$
$$-\omega_2 = -\omega_3 + \omega_1$$
$$-\omega_2 = \omega_1 - \omega_3$$

3! = 6 combinations & 3! = 6 combinations

$2 * 6 = 12$ different frequency relations

Each tensor has $3^3 = 27$ Cartesian components

For this condition there are $27 * 12 = 324$ $\chi^{(2)}$ elements

Double counting

- **Reality of the fields**
  - $\begin{matrix} \boldsymbol{E}^*(\omega_n) = \boldsymbol{E}(-\omega_n) \\ \boldsymbol{P}^*(\omega_n) = \boldsymbol{P}(-\omega_n) \end{matrix} \quad \Rightarrow \quad \chi^{(2)}_{ijk}(\omega_3;\omega_2,\omega_1)^* = \chi^{(2)}_{ijk}(-\omega_3;-\omega_2,-\omega_1)$
  - ½ of $\chi^{(2)}$ elements are lost
- **Intrinsic permutation symmetry**
  - $P_i^{(n)}(\omega_2) = \varepsilon_0 \sum_{jk} \chi^{(2)}_{ijk}(\omega_3;\omega_1,\omega_2) E_j(\omega_1) E_k(\omega_2) = \varepsilon_0 \sum_{jk} \chi^{(2)}_{ikj}(\omega_3;\omega_2,\omega_1) E_k(\omega_2) E_j(\omega_1)$
  - $\chi^{(2)}_{ijk}(\omega_3;\omega_1,\omega_2) = \chi^{(2)}_{ikj}(\omega_3;\omega_2,\omega_1)$

Loss-less material

- No absorption, non resonant process
- $\mathrm{Im}(\varepsilon) \to 0$
- $\mathrm{Im}(\chi_{ijk}^{(2)}) \to 0$
- All ω components can be interchanged as long as we also interchange the coresponding Cartesian components
- Example:

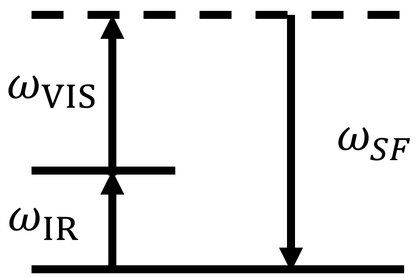
$$\overset{i}{\omega_3} = \overset{j}{\omega_1} + \overset{k}{\omega_2}$$

$$\chi_{ijk}^{(2)}(\omega_3; \omega_1, \omega_2) = \chi_{jki}^{(2)}(-\omega_1; \omega_2, -\omega_3) = \chi_{jki}^{(2)} \quad {}^{*} = \chi_{jki}^{(2)}(\omega_1; -\omega_2, \omega_3\Big)$$

$$-\overset{j}{\omega_1} = \overset{k}{\omega_2} - \overset{i}{\omega_3}$$

Kleinman's symmetry

- Dispersion $\chi^{(n)}(\omega)$ can be negligible
- Frequency components are close to each other and the material is transparent
- $\chi^{(2)}_{ijk}(\omega_3;\omega_1,\omega_2) = \chi^{(2)}_{ijk}(\omega_1;\omega_2,\omega_3\big)$
- Now we have a full decoupling of frequencies and polarization indices
- Example: SF spectroscopy



VIS and SF are off-resonance, close in frequency

$$\chi^{(2)}_{ijk}(\omega_{SF};\omega_{VIS},\omega_2) = \chi^{(2)}_{jik}(\omega_{SF};\omega_{VIS},\omega_2\big)$$

- From 27 $\chi^{(2)}$ elements (3^3)
- $\chi_{zxx} = \chi_{xzx} = \chi_{xxz}, \ldots$
  $\chi_{xyz} = \chi_{xzy} = \chi_{yzx} = \chi_{yxz} = \chi_{zxy} = \chi_{zyx}$
- 10 independent non zero elements (xxx, yyy, zzz, xyy, xzz, yxx, yzz, zxx, zyy, xyz)

Contracted notation for $\chi^{(2)}$

- Used for crystal and for SHG where two frequencies are indistinguishable in terms of their NLO response
- $d_{ijk} = \frac{1}{2}\chi_{ijk}^{(2)}$
- $d_{il} = 111, 122, 133, 123 = 132, 131 = 113, 112 = 121$

l *i1* *i2* *i3* *i4* *i5* *i6*