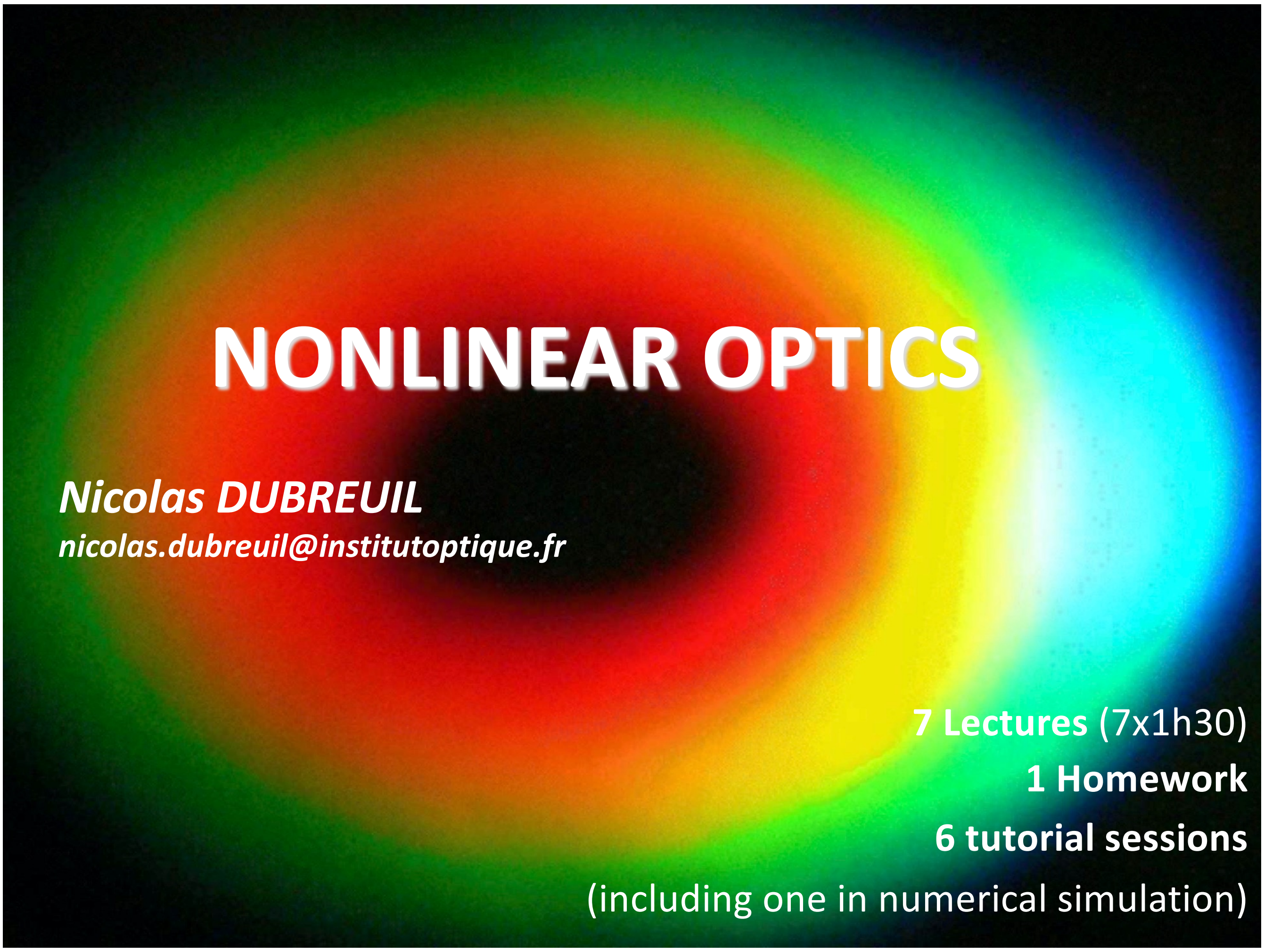


Lecture 3 /7 : learning outcomes

By the end of this lecture, students will know...

• the constitutive relations of nonlinear optics ($D = \epsilon_0 E + P$ and $P = \epsilon_0 \chi^{(1)} E + \epsilon_0 \chi^{(2)} EE + \epsilon_0 \chi^{(3)} EEE + \cdots$) (K1)
• the basic properties of nonlinear susceptibility tensors (K4)

By the end of this course, students will be skilled at ...

- Manipulating the nonlinear susceptibility tensor components and, with given incident fields, calculate the components of nonlinear polarisation vector

Nonlinear Optics

- **First descriptions**
  - **Expression of the macroscopic polarization in terms of a power series in the field strength :**

$$\mathcal{P}(t) = \chi_1 \mathcal{E}(t) + \chi_2 \mathcal{E}(t)\mathcal{E}(t) + \chi_3 \mathcal{E}(t)\mathcal{E}(t)\mathcal{E}(t) + \cdots,$$

  - **Origin of the nonlinearities : classical anharmonic oscillator (classical model)**

- **What are the limits ?**
  - **the response of material was described by a scalar quantity**
  - **the response time of the material was assumed to be infinitely short (instantaneous response)**

Lecture 3 - Content

- **Constitutive relations on nonlinear optics**
  - Derivation of the impulse response of a time invariant and causal system in LINEAR and NONLINEAR regimes

- **Nonlinear susceptibility tensors**
  - Definition
  - Basic properties

Field notation

We assume that the electric field vector can be expressed as a plane wave (or as a projection of plane waves, i.e through a Fourier transformation) :

$$\boldsymbol{\mathcal{E}}(t) = \boldsymbol{E}(\omega)e^{\imath(-\omega t+\boldsymbol{k}\cdot\boldsymbol{r})} + \boldsymbol{E}^*(\omega)e^{-\imath(-\omega t+\boldsymbol{k}\cdot\boldsymbol{r})} \quad \text{With :} \quad \boldsymbol{E}(\omega) = \left| \begin{array}{l} E_i(\omega) \\ E_j(\omega) \\ E_k(\omega) \end{array} \right.$$

$$\underline{\boldsymbol{\mathcal{E}}(t)} = E(\omega)e^{\imath(-\omega t+\boldsymbol{k}\cdot\boldsymbol{r})}\boldsymbol{e} + CC$$

Purely REAL quantity

Polarization state

Notation :
$$\boxed{E^*(\omega) = E(-\omega)}$$

Similarly for the macroscopic polarization :

$$\underline{\boldsymbol{\mathcal{P}}(t)} = P(\omega)e^{\imath(-\omega t+\boldsymbol{k}\cdot\boldsymbol{r})}\boldsymbol{e} + P^*(\omega)e^{-\imath(-\omega t+\boldsymbol{k}\cdot\boldsymbol{r})}\boldsymbol{e}$$

Purely REAL quantity

Notation :
$$\boxed{P^*(\omega) = P(-\omega)}$$

Pulse response in LINEAR regime

Description of the propagation of an electromagnetic field E(t) through a linear medium :

- LINEAR MEDIUM = LINEAR FILTER
- TIME INVARIANT & CAUSAL linear system (+LOCAL Response)

$$\underset{\boldsymbol{\mathcal{E}}(t)}{\text{Excitation}} \longrightarrow \boxed{\text{Linear Medium}} \longrightarrow \underset{\boldsymbol{\mathcal{P}}(t)}{\text{Response}}$$

$$\boldsymbol{\mathcal{P}}(t) = \epsilon_0 \int_{-\infty}^{+\infty} \underline{\underline{T}}^{(1)}(t,\tau)\boldsymbol{\mathcal{E}}(\tau)d\tau$$

$\underline{\underline{T}}^{(1)}(t,\tau)$: Impulse response of the Medium : general case of an anisotropic material, it is a tensor of rank 2 (i.e. 3x3 matrix)

Properties :

-***E****(t)* and ***P****(t)* are purely real quantities, same for $\underline{T}^{(1)}(t,T)$

- time invariant system :

$$\underline{\underline{T}}^{(1)}(t,\tau) = \underline{\underline{R}}^{(1)}(t-\tau)$$

$$\begin{aligned} \boldsymbol{P}(t) &= \epsilon_0 \int \underline{\underline{R}}^{(1)}(t-\tau)\boldsymbol{E}(\tau)d\tau, \\ &= \epsilon_0 \int \underline{\underline{R}}^{(1)}(\tau)\boldsymbol{E}(t-\tau)d\tau \end{aligned}$$

-Causal system implies that $\underline{\underline{R}}^{(1)}(\tau) = 0 \text{ for } \tau < 0$

Linear susceptibility

Convention for the Fourier Transform :

$$\boldsymbol{\mathcal{E}}(t) = \int \boldsymbol{E}(\omega) e^{-\imath\omega t} d\omega$$

$$\boldsymbol{E}(\omega) = \frac{1}{2\pi} \int \boldsymbol{\mathcal{E}}(t) e^{+\imath\omega t} dt$$

$$\begin{aligned}\boldsymbol{\mathcal{P}}(t) &= \epsilon_0 \int \underline{\underline{R}}^{(1)}(t-\tau)\boldsymbol{\mathcal{E}}(\tau)d\tau, \\ &= \epsilon_0 \int \underline{\underline{R}}^{(1)}(\tau)\boldsymbol{\mathcal{E}}(t-\tau)d\tau\end{aligned}
\xrightarrow{\text{Fourier Transform}}
\boxed{\boldsymbol{P}(\omega) = \epsilon_0 \underbrace{\underline{\underline{\chi}}^{(1)}(\omega)}\boldsymbol{E}(\omega)}$$

Linear Susceptibility : Fourier transform of the pulse response

$$\underline{\underline{\chi}}^{(1)}(\omega) = 2\pi \,\mathrm{TF}\left[\underline{\underline{R}}^{(1)}(t)\right]$$

Properties :

- Reality of the function $\underline{\underline{R}}^{(1)}(t)$ ➡ $\underline{\underline{\chi}}^{(1)}(\omega)^\star = \underline{\underline{\chi}}^{(1)}(-\omega)$

- The causality property enables to derive the Kramers-Kroning relations that relates the real and imaginary parts of the linear susceptibility

Nonlinear pulse response

Propagation of an electromagnetic field E(t) through a NONlinear medium :

NONLINEAR MEDIUM = TIME INVARIANT & CAUSAL system (+ LOCAL Response)

Excitation $\boldsymbol{\mathcal{E}}(t)$ → NonLinear Medium → Response

$$\boldsymbol{\mathcal{P}}(t) = \boldsymbol{\mathcal{P}}^{(1)}(t) + \boldsymbol{\mathcal{P}}^{(2)}(t) + \boldsymbol{\mathcal{P}}^{(3)}(t) + \cdots$$

$$\boldsymbol{\mathcal{P}}(t) = \epsilon_0 \int_{-\infty}^{+\infty} \underline{\underline{T}}^{(1)}(t,\tau)\boldsymbol{\mathcal{E}}(\tau)d\tau$$ Linear Macroscopic Polarization

$$\boxed{\boldsymbol{\mathcal{P}}^{(2)}(t) = \epsilon_0 \int\int \underbrace{\underline{\underline{\underline{T}}}^{(2)}(t;\tau_1,\tau_2)}\boldsymbol{\mathcal{E}}(\tau_1)\boldsymbol{\mathcal{E}}(\tau_2)d\tau_1 d\tau_2}$$

2nd order nonlinear Macroscopic Polarization

2nd order nonlinear impulse response = 3rd order tensor (in order to fully describe the quadratic dependance of the 2nd order nonlinear polarization)

Expression of the ith component : $\mathcal{P}_i^{(2)}(t) = \epsilon_0 \sum_{(j,k)} \int\int T_{ijk}^{(2)}(t;\tau_1,\tau_2)\mathcal{E}_j(\tau_1)\mathcal{E}_k(\tau_2)d\tau_1 d\tau_2$

Component *ijk* of the tensor

Constitutive relations : nonlinear pulse response

nth order nonlinear pulse response :

$$\mathcal{P}^{(n)}(t) = \epsilon_0 \int\int \cdots \int \underline{\underline{R}}^{(n)}(t;\tau_1,\tau_2,\cdots,\tau_n)\boldsymbol{\mathcal{E}}(t-\tau_1)\boldsymbol{\mathcal{E}}(t-\tau_2)\cdots\boldsymbol{\mathcal{E}}(t-\tau_n)d\tau_1 d\tau_2\cdots d\tau_n$$

PROPERTIES of the nonlinear pulse response :

Symmetry condition: A tensor can be expressed as the summation of a symmetric and an antisymmetric tensor

$$T^{(2)}_{ijk}(t;\tau_1,\tau_2) = S^{(2)}_{ijk}(t;\tau_1,\tau_2) + A^{(2)}_{ijk}(t;\tau_1,\tau_2)$$

$$S^{(2)}_{ijk} = \frac{1}{2}\left[T^{(2)}_{ijk}(t;\tau_1,\tau_2) + T^{(2)}_{ikj}(t;\tau_2,\tau_1)\right]$$

$$A^{(2)}_{ijk} = \frac{1}{2}\left[T^{(2)}_{ijk}(t;\tau_1,\tau_2) - T^{(2)}_{ikj}(t;\tau_2,\tau_1)\right]$$

Substitution into $P^{(2)}_i(t) = \epsilon_0 \sum_{(j,k)} T^{(2)}_{ijk}(t;\tau_1,\tau_2)E_j(\tau_1)E_k(\tau_2)d\tau_1 d\tau_2$

Shows that

$$\boxed{T^{(2)}_{ijk}(t;\tau_1,\tau_2) = T^{(2)}_{ikj}(t;\tau_2,\tau_1)}$$

Conclusion : the nonlinear pulse response tensor is SYMMETRIC

Constitutive relations : nonlinear pulse response

PROPERTIES of the nonlinear pulse response :

Time invariant response property : $\underline{\underline{T}}^{(2)}(t;\tau_1,\tau_2) = \underline{\underline{R}}^{(2)}(t-\tau_1, t-\tau_2)$

$$\begin{aligned}\mathcal{P}^{(2)}(t) &= \epsilon_0 \int\int \underline{\underline{R}}^{(2)}(t-\tau_1,t-\tau_2)\boldsymbol{\mathcal{E}}(\tau_1)\boldsymbol{\mathcal{E}}(\tau_2)d\tau_1 d\tau_2,\\ &= \epsilon_0 \int\int \underline{\underline{R}}^{(2)}(\tau_1,\tau_2)\boldsymbol{\mathcal{E}}(t-\tau_1)\boldsymbol{\mathcal{E}}(t-\tau_2)d\tau_1 d\tau_2.\end{aligned}$$

Causal system: $\underline{\underline{R}}^{(2)}(\tau_1,\tau_2) = 0 \text{ for } \tau_1 < 0 \text{ and } \tau_2 < 0$

Real function: The field vectors are real quantities, which imply the reality of the nonlinear pulse response.

Constitutive relations : nonlinear susceptibility

Case of the 2nd order nonlinear susceptibility :

$$\boxed{\boldsymbol{P}^{(2)}(\omega=\omega_1+\omega_2)=\epsilon_0\,\underline{\underline{\chi}}^{(2)}(\omega=\omega_1+\omega_2;\omega_1,\omega_2)\boldsymbol{E}(\omega_1)\boldsymbol{E}(\omega_2)}$$

$$\begin{aligned}\underline{\underline{\chi}}^{(2)}(\omega_s;\omega_1,\omega_2) &= \int\int \underline{\underline{R}}^{(2)}(\tau_1,\tau_2)e^{\imath(\omega_1\tau_1+\omega_2\tau_2)}d\tau_1 d\tau_2\\ &= (2\pi)^2\mathrm{TF}\left[\underline{\underline{R}}^{(2)}(\tau_1,\tau_2)\right].\end{aligned}$$

$$\boxed{P_i^{(2)}(\omega_1+\omega_2)=\epsilon_0\sum_{jk}\chi_{ijk}^{(2)}(\omega_1+\omega_2;\omega_1,\omega_2)E_j(\omega_1)E_k(\omega_2)}$$

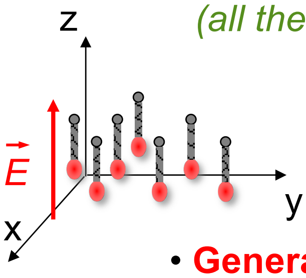
$$\boldsymbol{P}(\omega=\omega_p+\omega_q)=\epsilon_0\sum_{(pq)}\underline{\underline{\chi}}^{(2)}(\omega=\omega_p+\omega_q;\omega_p,\omega_q)\boldsymbol{E}(\omega_p)\boldsymbol{E}(\omega_q)$$

Nonlinear susceptibility tensor

Case of the nonlinear interaction of 2 waves @ ω_1 and ω_2 in a 2nd order NL medium :

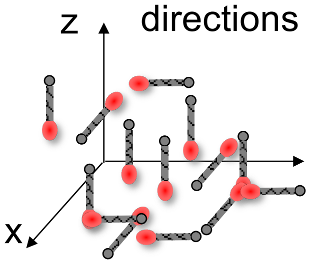
•Classical anharmonic oscillator : ***scalar* expression** of the polarization @ $\omega=\omega_1+\omega_2$

(all the dipoles are supposed identically oriented along the linear polarization state of the applied field):



$$P(\omega_1+\omega_2)=\epsilon_0\chi^{(2)}(\omega_1,\omega_2)E(\omega_1)E(\omega_2)$$

• **General description :** the array of dipoles are oriented along the 3 directions x,y et z + different oscillator parameters for each direction



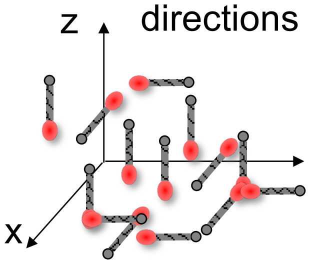
General relation :

$$P_i(\omega_1+\omega_2)=\epsilon_0\sum_{jk}\chi_{ijk}^{(2)}(\omega_1+\omega_2;\omega_1,\omega_2)E_j(\omega_1)E_k(\omega_2)$$

Nonlinear susceptibility tensor

Case of the nonlinear interaction of 2 waves @ ω_1 and ω_2 in a 2^nd^ order NL medium :

- **General description :** the array of dipoles are oriented along the 3 directions x,y et z + different oscillator parameters for each direction



General relation :

$$P_i(\omega_1+\omega_2) = \epsilon_0 \sum_{jk} \chi^{(2)}_{ijk}(\omega_1+\omega_2;\omega_1,\omega_2)E_j(\omega_1)E_k(\omega_2)$$

Vector / Tensor notation :

$$\boldsymbol{P}(\omega_1+\omega_2) = \epsilon_0 \underline{\underline{\underline{\chi}}}^{(2)}(\omega_1+\omega_2;\omega_1,\omega_2)\boldsymbol{E}(\omega_1)\boldsymbol{E}(\omega_2)$$

Vector

Tensor of rank 3

Vectors

Nonlinear susceptibility tensor

To conclude

- The response of a material subject to the excitation by an electromagnetic field is given by the **macroscopic polarization**
- In the electric dipole approximation (which consists in neglecting the quadripole term in the constitutive relation), the **polarization term is developed in power series expansion of the electric field:**

$$\boldsymbol{\mathcal{P}}(t) = \boldsymbol{\mathcal{P}}^{(1)}(t) + \boldsymbol{\mathcal{P}}^{(2)}(t) + \boldsymbol{\mathcal{P}}^{(3)}(t) + \cdots$$

- **Relation with the nonlinear impulse response of the material :**

$$\begin{aligned}\boldsymbol{\mathcal{P}}^{(2)}(t) &= \int \boldsymbol{P}^{(2)}(\omega)e^{-\imath\omega t}d\omega \\ &= \epsilon_0 \int\int \underline{\underline{\underline{R}}}^{(2)}(\tau_1,\tau_2)\boldsymbol{\mathcal{E}}(t-\tau_1)\boldsymbol{\mathcal{E}}(t-\tau_2)d\tau_1 d\tau_2\end{aligned}$$

vector

Tensor of rank 3 (impulse response)

vectors

Nonlinear susceptibility tensor

To conclude

- **Relation with the nonlinear impulse response of the material :**

$$\begin{aligned}\boldsymbol{\mathcal{P}}^{(2)}(t) &= \int \boldsymbol{P}^{(2)}(\omega)e^{-\imath\omega t}d\omega \\ &= \epsilon_0 \int\int \underline{\underline{R}}^{(2)}(\tau_1,\tau_2)\boldsymbol{\mathcal{E}}(t-\tau_1)\boldsymbol{\mathcal{E}}(t-\tau_2)d\tau_1 d\tau_2\end{aligned}$$

- **The nonlinear susceptibility is proportional to the FOURIER TRANSFORM of the impulse response of the material**

$$\begin{aligned}\underline{\underline{\chi}}^{(2)}(\omega_1+\omega_2;\omega_1,\omega_2) &= \int\int \underline{\underline{R}}^{(2)}(\tau_1,\tau_2)e^{\imath(\omega_1\tau_1+\omega_2\tau_2)}d\tau_1 d\tau_2. \\ &= (2\pi)^2\mathrm{TF}\left[\underline{\underline{R}}^{(2)}(\tau_1,\tau_2)\right].\end{aligned}$$

- **Constitutive relation of nonlinear optics =** $\boxed{\boldsymbol{P}^{(2)}(\omega=\omega_1+\omega_2)=\epsilon_0\,\underline{\underline{\chi}}^{(2)}(\omega=\omega_1+\omega_2;\omega_1,\omega_2)\boldsymbol{E}(\omega_1)\boldsymbol{E}(\omega_2)}$

This constitutive relation means that the interaction of two waves at ω_1 and ω_2 in a second order nonlinear medium generates a polarization term at $\omega=\omega_1+\omega_2$. Notice that the frequency arguments can take either positive or negative values, which comes from the assumption for the electric field $\boldsymbol{\mathcal{E}}(t)$ to be a purely real quantity

Nonlinear susceptibility tensor

To conclude

- **Constitutive relation of nonlinear optics =** $\boxed{\boldsymbol{P}^{(2)}(\omega=\omega_1+\omega_2)=\epsilon_0\,\underline{\underline{\chi}}^{(2)}(\omega=\omega_1+\omega_2;\omega_1,\omega_2)\boldsymbol{E}(\omega_1)\boldsymbol{E}(\omega_2)}$

This constitutive relation means that the interaction of two waves at ω_1 and ω_2 in a second order nonlinear medium generates a polarization term at $\omega=\omega_1+\omega_2$. Notice that the frequency arguments can take either positive or negative values, which comes from the assumption for the electric field $\boldsymbol{\mathcal{E}}(t)$ to be a purely real quantity (see (1.7)).

Each vectorial component of the polarization can be expressed in terms the tensorial components $\chi^{(2)}_{ijk}(\omega=\omega_1+\omega_2;\omega_1,\omega_2)$ and the electric field components $E_{i,j,k}(\omega)$:

$$P_i^{(2)}(\omega_1+\omega_2)=\epsilon_0\sum_{jk}\chi^{(2)}_{ijk}(\omega_1+\omega_2;\omega_1,\omega_2)E_j(\omega_1)E_k(\omega_2).$$

This relation might also require to sum over the frequency components :

$$P_i(\omega=\omega_p+\omega_q)=\epsilon_0\sum_{jk}\sum_{(pq)}\chi^{(2)}_{ijk}(\omega=\omega_p+\omega_q;\omega_p,\omega_q)E_j(\omega_p),E_k(\omega_q).$$

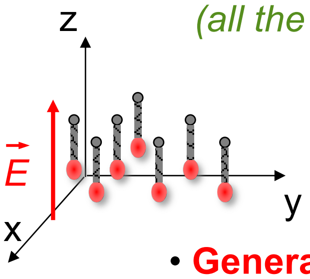
$$\boldsymbol{P}(\omega=\omega_p+\omega_q)=\epsilon_0\sum_{(pq)}\underline{\underline{\chi}}^{(2)}(\omega=\omega_p+\omega_q;\omega_p,\omega_q)\boldsymbol{E}(\omega_p)\boldsymbol{E}(\omega_q)$$

Nonlinear susceptibility tensor - Definition

Case of the nonlinear interaction of 2 waves @ ω_1 and ω_2 in a 2nd order NL medium :

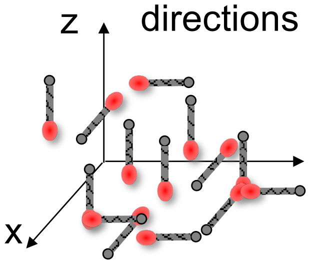
•Classical anharmonic oscillator : ***scalar* expression** of the polarization @ $\omega=\omega_1+\omega_2$

(all the dipoles are supposed identically oriented along the linear polarization state of the applied field):



$$P(\omega_1+\omega_2)=\epsilon_0\chi^{(2)}(\omega_1,\omega_2)E(\omega_1)E(\omega_2)$$

• **General description :** the array of dipoles are oriented along the 3 directions x,y et z + different oscillator parameters for each direction



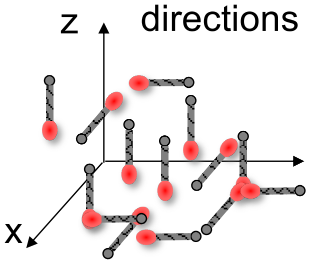
General relation :

$$P_i(\omega_1+\omega_2)=\epsilon_0\sum_{jk}\chi^{(2)}_{ijk}(\omega_1+\omega_2;\omega_1,\omega_2)E_j(\omega_1)E_k(\omega_2)$$

Nonlinear susceptibility tensor - Definition

Case of the nonlinear interaction of 2 waves @ ω_1 and ω_2 in a 2nd order NL medium :

• **General description :** the array of dipoles are oriented along the 3 directions x,y et z + different oscillator parameters for each direction



General relation :

$$P_i(\omega_1+\omega_2)=\epsilon_0\sum_{jk}\chi^{(2)}_{ijk}(\omega_1+\omega_2;\omega_1,\omega_2)E_j(\omega_1)E_k(\omega_2)$$

Vector / Tensor notation :

$$\boldsymbol{P}(\omega_1+\omega_2)=\epsilon_0\underline{\underline{\underline{\chi}}}^{(2)}(\omega_1+\omega_2;\omega_1,\omega_2)\boldsymbol{E}(\omega_1)\boldsymbol{E}(\omega_2)$$

Vector

Tensor of rank 3

Vectors

Nonlinear susceptibility tensor - Definition

✓ **2nd order NL susceptibility :**

= tensor of rank 3 $\underline{\underline{\underline{\chi}}}^{(2)}$

It contains 9x 3 = 27 components

Comment : *Each tensor is defined for a set of frequencies.*
The value of the components of the tensor depends on the frequencies (in a general manner) !!!

- **General expression of the 2nd order NL polarization :**

$$\boldsymbol{P}(\omega=\omega_p+\omega_q)=\epsilon_0\sum_{(pq)}\underline{\underline{\underline{\chi}}}^{(2)}(\omega=\omega_p+\omega_q;\omega_p,\omega_q)\boldsymbol{E}(\omega_p)\boldsymbol{E}(\omega_q)$$

Expression of the i^{th} component :

$$P_i(\omega=\omega_p+\omega_q)=\epsilon_0\sum_{jk}\sum_{(pq)}\chi^{(2)}_{ijk}(\omega=\omega_p+\omega_q;\omega_p,\omega_q)E_j(\omega_p)E_k(\omega_q)$$

Nonlinear susceptibility tensor - Definition

✓ **3rd order NL susceptibility :**

= tensor of rank 4 $\underline{\underline{\underline{\chi}}}^{(3)}$

81 components !!!!

- **General expression of the 3rd order NL polarization :**

$$P_i(\omega=\omega_p+\omega_q+\omega_r)=\epsilon_0\sum_{jkl}\sum_{(pqr)}\chi^{(3)}_{ijkl}(\omega=\omega_p+\omega_q+\omega_r;\omega_p,\omega_q,\omega_r)E_j(\omega_p)E_k(\omega_q)E_l(\omega_r)$$

Expression of the i^{th} component :

$$\boldsymbol{P}(\omega=\omega_p+\omega_q+\omega_r)=\epsilon_0\sum_{(pqr)}\underline{\underline{\underline{\chi}}}^{(3)}(\omega=\omega_p+\omega_q+\omega_r;\omega_p,\omega_q,\omega_r)\boldsymbol{E}(\omega_p)\boldsymbol{E}(\omega_q)\boldsymbol{E}(\omega_r)$$

✓ **Nth order NL susceptibility**

… just have fun !!

Properties of NL susceptibilities

Nonlinear susceptibilities = Tensor

$$\omega_1,\ \omega_2 \longrightarrow \boxed{\underline{\underline{\chi}}^{(2)}} \longrightarrow \omega_3 = \omega_1 + \omega_2$$

$$\underline{\underline{\chi}}^{(2)}$$

Complete description of the waves interaction (3 waves in this case) requires the determination of :

$$\vec{P}(\omega_1), \vec{P}(\omega_2), \vec{P}(\omega_3) \qquad \vec{P}(-\omega_1), \vec{P}(-\omega_2), \vec{P}(-\omega_3)$$

$\underline{\underline{\chi}}^{(2)}(\omega_1; \omega_3, -\omega_2)$ $\quad$ $\underline{\underline{\chi}}^{(2)}(\omega_2; \omega_3, -\omega_1)$ $\quad$ ... $\quad$ ... $\quad$ ... $\quad$ ...

$\underline{\underline{\chi}}^{(2)}(\omega_1; -\omega_2, \omega_3)$ $\quad$ $\underline{\underline{\chi}}^{(2)}(\omega_2; -\omega_1, \omega_3)$ $\quad$ ... $\quad$ ... $\quad$ ... $\quad$ ...

12 tensors = 12 x 27 = 324 components !!!

Properties of NL susceptibilities

- ***Reality of the fields***

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

- ***Intrinsic Permutation Symmetry***

The quantities : $\chi_{ijk}^{(2)}(\omega_3; \omega_1, \omega_2) E_j(\omega_1) E_k(\omega_2)$

and $\chi_{ikj}^{(2)}(\omega_3; \omega_2, \omega_1) E_k(\omega_2) E_j(\omega_1)$ are numerically equal

Consequence

$$\rightarrow \quad \chi_{ijk}^{(2)}(\omega_3 = \omega_1 + \omega_2; \omega_1, \omega_2) = \chi_{ikj}^{(2)}(\omega_3 = \omega_1 + \omega_2; \omega_2, \omega_1)$$

- ***Lossless media***

Expression of χ^{NL} is a purely real quantity

Verification : in the case of the classical oscillator model discussed in ch1, since $\omega << \omega_0$

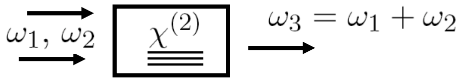
Properties of NL susceptibilities

- ***Degeneracy Factor***

Determination of *P(ω)* : summation over field frequencies in interaction and for which $\omega = \omega_1 + \omega_2 + \omega_3 + \cdots$

Due to intrinsic permutation $\longrightarrow$ simplification occurs

Example : Sum-Frequency generation



$$P_i(\omega_3) = \epsilon_0 \sum_{jk} \left[\chi^{(2)}_{ijk}(\omega_3;\omega_1,\omega_2) E_j(\omega_1) E_k(\omega_2) + \chi^{(2)}_{ijk}(\omega_3;\omega_2,\omega_1) E_j(\omega_2) E_k(\omega_1) \right]$$

Intrinsic permutation $\longrightarrow$ $P_i(\omega_3) = 2\epsilon_0 \sum_{jk} \chi^{(2)}_{ijk}(\omega_3;\omega_1,\omega_2) E_j(\omega_1) E_k(\omega_2)$

Properties of NL susceptibilities

- ***Degeneracy Factor***

- 2nd order NL Polarization expression

$$P_i(\omega_3) = D^{(2)} \epsilon_0 \sum_{jk} \chi^{(2)}_{ijk}(\omega_3;\omega_1,\omega_2) E_j(\omega_1) E_k(\omega_2)$$

Degeneracy factor

- $D^{(2)} = 1$ in the case of one distinct field $(\omega_1 = \omega_2)$,
- $D^{(2)} = 2$ in the case of 2 distinct fields $(\omega_1 \neq \omega_2)$.

- 3rd order NL Polarization expression

$$P_i(\omega_4 = \omega_1 + \omega_2 + \omega_3) = D^{(3)} \epsilon_0 \sum_{jkl} \chi^{(3)}_{ijkl}(\omega_4;\omega_1,\omega_2,\omega_3) E_j(\omega_1) E_k(\omega_2) E_l(\omega_3$$

Degeneracy factor

- $D^{(3)} = 1$ in the case of one distinct field $(\omega_1 = \omega_2 = \omega_3)$,
- $D^{(3)} = 3$ in the case of 2 distinct fields $(\omega_1 = \omega_2 \neq \omega_3)$,
- $D^{(3)} = 3! = 6$ in the case of 3 distinct fields $(\omega_1 \neq \omega_2 \neq \omega_3)$.

Properties of NL susceptibilities

• *Kleinman Symmetry - Lossless Media*

Lossless media : no exchange of energy with the nonlinear medium

$$\chi^{(2)}_{ijk}(\omega_3=\omega_1+\omega_2;\omega_1,\omega_2)=\chi^{(2)}_{jki}(-\omega_1=\omega_2-\omega_3;\omega_2,-\omega_3)=\chi^{(2)}_{kji}(-\omega_2=\omega_1-\omega_3;\omega_1,-\omega_3).$$

(See Tutorial 1)

Simultaneous permutations of the indices with the frequency arguments

Far from any material resonance, χ^{NL} does not depend on frequencies **Consequence :**

Permutation of the indices without permuting frequencies

$$\chi^{(2)}_{ijk}(\omega_3=\omega_1+\omega_2;\omega_1,\omega_2)=\chi^{(2)}_{jki}(\omega_3=\omega_1+\omega_2;\omega_1,\omega_2)=\chi^{(2)}_{kji}(\omega_3=\omega_1+\omega_2;\omega_1,\omega_2)$$

+ intrinsic permutation

Full permutation of the indices, without permuting the frequencies

Spatial Symmetries

Spatial symmetry properties of the nonlinear material : reduction of the number of independent components

• Example : media inside which the directions x and y are similar (from th point of view of its NL response)

$\chi^{(2)}_{zxx}=\chi^{(2)}_{zyy}$ (for instance) → Strong reduction of the numbers of independent components

• **Important example : Centre-symmetric material**

2nd order nonlinear susceptibility vanishes (i.e silica...)

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

(generalization : $2N^{th}$ order)

Contracted notation d_{eff}

When the **Kleinman symmetry** condition is valid
Or
For **2nd harmonic generation** process

$$d_{ijk} = \frac{1}{2}\chi^{(2)}_{ijk}$$

Permutation symmetry of the last two indices

Contraction notation of the last two indices

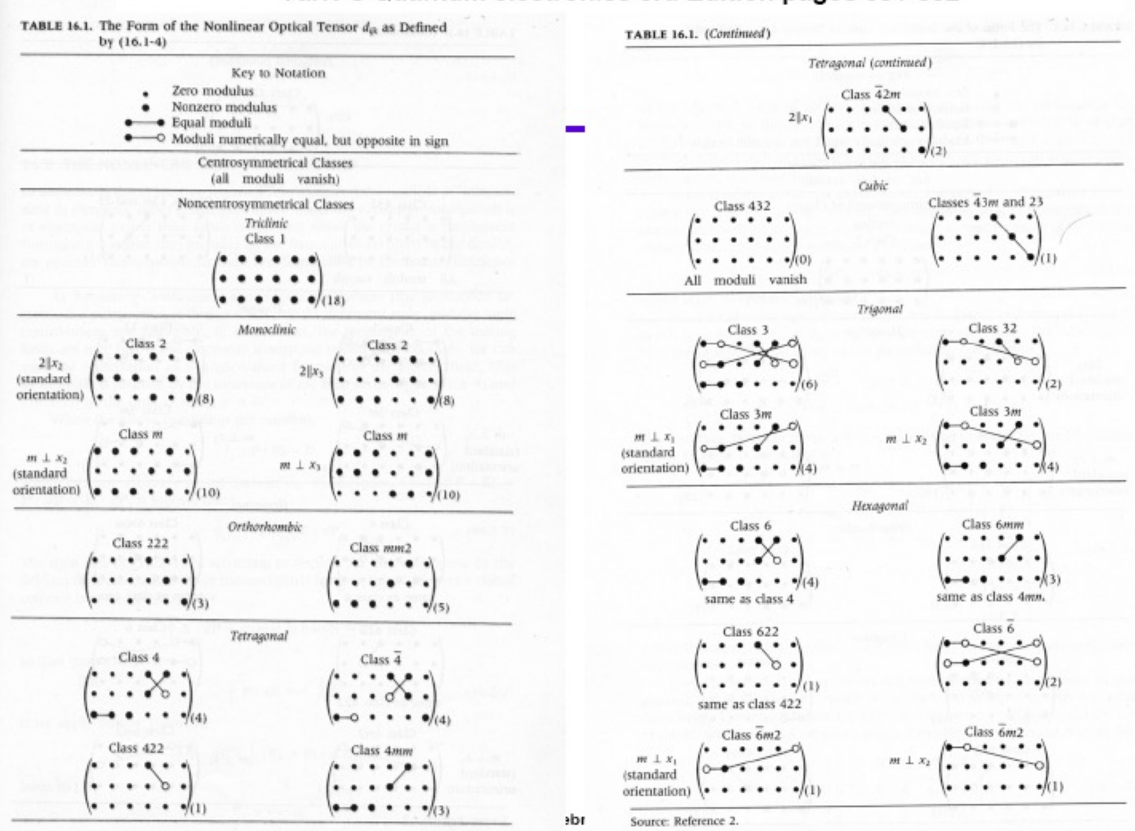
$$\begin{array}{c|cccccc} jk & 11 & 22 & 33 & 23 \text{ ou } 32 & 13 \text{ ou } 31 & 12 \text{ ou } 21 \\ l & 1 & 2 & 3 & 4 & 5 & 6 \end{array}$$

$$d_{il} = \begin{bmatrix} d_{11} & d_{12}\cdots & d_{16} \\ d_{21} & \cdots & d_{26} \\ d_{31} & \cdots & d_{36} \end{bmatrix}$$

Matrix with 6x3 components

Spatial Symmetries

Yariv's Quantum electronics 3rd Edition pages 381-382



Spatial Symmetries

EXAMPLE : KDP crystal – See the learning activity on eCampus

Point group $4\bar{2}m$ - 3 nonzero coefficient, 2 numerically equal coefficents :

$$\begin{bmatrix} 0 & 0 & 0 & d_{14} & 0 & 0 \\ 0 & 0 & 0 & 0 & d_{14} & 0 \\ 0 & 0 & 0 & 0 & 0 & d_{36} \end{bmatrix}$$

2ω generation : Determination of $\vec{P}(2\omega)$

$$\begin{aligned} P_x(2\omega) &= 4\epsilon_0 d_{14} E_y(\omega) E_z(\omega) \\ P_y(2\omega) &= 4\epsilon_0 d_{14} E_x(\omega) E_z(\omega) \\ P_z(2\omega) &= 4\epsilon_0 d_{36} E_x(\omega) E_y(\omega) \end{aligned}$$

Student activities

To complete, read the lecture notes : sections 2.2 and 2.3

+ Complete two short tests on eCampus by next Monday (27 Nov.)