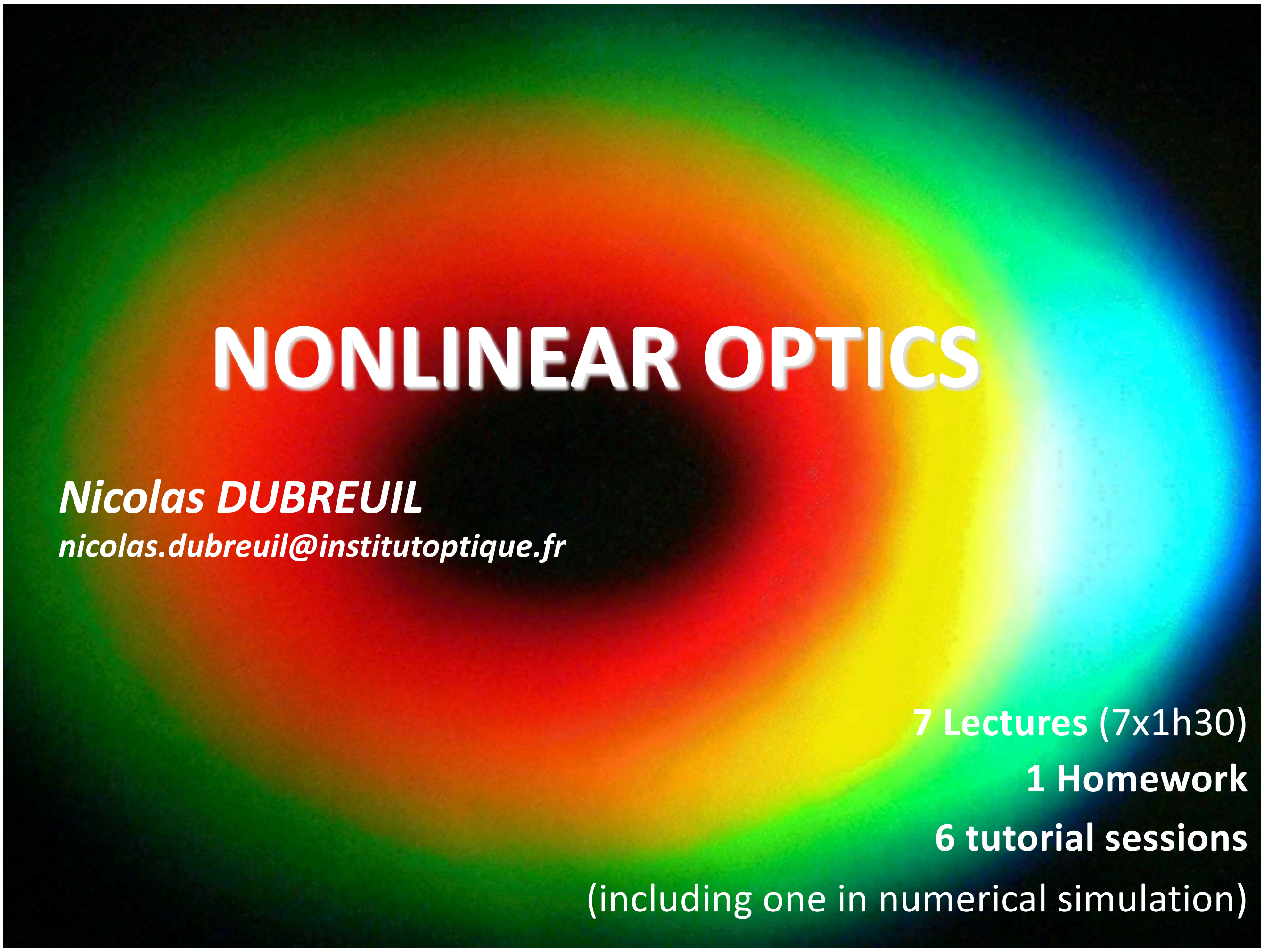

NONLINEAR OPTICS

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7 Lectures (7x1h30)
1 Homework
6 tutorial sessions
(including one in numerical simulation)

Lecture 1/7 : learning outcomes

By the end of this lecture, you will be able to ...
• cite nonlinear effects that arise in a 2nd and 3rd order nonlinear materials (K2)
• provide a classical description for the origin of the nonlinear susceptibilities (K3)

By the end of this lecture, you will start to understand ...
• the capability of light matter interactions in modifying light properties : frequency generation, optical rectification… (Q1)
• how a perturbative approache enables in describing and deriving a NON LINEAR problem in physics (Q2)
• the link between the microscopic and macroscopic terms in Maxwell's equations (induced dipole, macroscopic polarization and fields) (Q3)

Lecture 2 /7 : learning outcomes

By the end of this two lectures, you 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 lecture, you will be skilled at...

- deriving and solving the nonlinear wave equation in a parametric situation under the undepleted pump approximation (S3)

By the end of this lecture, you will understand ...

- Nonlinear effects are subject to phase matching conditions (U5)

Lecture 2 - Content

- **Field notation**

- **Introduction to nonlinear susceptibility tensors**

- **Nonlinear wave equation: application to SHG & Phase matching condition**

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) :

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

With : $\boldsymbol{E}(\omega) = \begin{vmatrix} E_i(\omega) \\ E_j(\omega) \\ E_k(\omega) \end{vmatrix}$

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

Purely REAL quantity

Polarization state

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

Similarly for the macroscopic polarization :

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

Purely REAL quantity

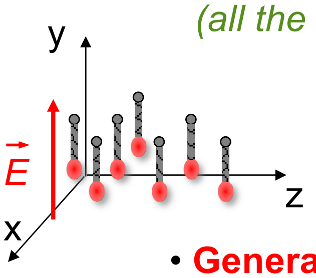
Notation : $P^*(\omega) = P(-\omega)$

Nonlinear susceptibility tensor - Definition

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

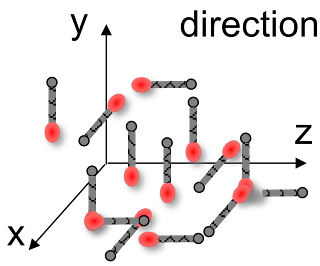
•Classical anharmonic oscillator : ***scalar* expression** of the polarization @ $\omega=\omega_1+\omega_2$

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



$$P_y(\omega_1+\omega_2) = \epsilon_0\chi^{(2)}_{yyy}(\omega_1,\omega_2)E_y(\omega_1)E_y(\omega_2)$$

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



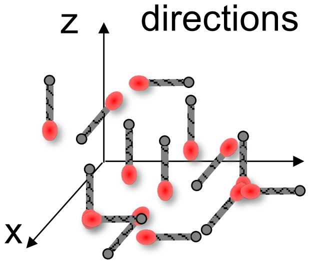
General relation :

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

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 (P) — Tensor of rank 3 ($\chi^{(2)}$) — Vectors (E)

Lecture 2 - Content

- Field notation
  - Introduction to nonlinear susceptibility tensors
- **Nonlinear wave equation: application to SHG & Phase matching condition**

NONLINEAR OPTICS

Nonlinear wave equation: application to SHG & Phase matching condition

- **Maxwell's equations**
- **Nonlinear wave equation in a isotropic material**
  - ➢ Application : Second Harmonic Generation (SHG)
  - ➢ Discussion about the phase matching condition
- Propagation in a linear anisotropic material
- Stationary nonlinear wave equation
- Phase Matching considerations

NONLINEAR OPTICS

Nonlinear wave equation: application to SHG & Phase matching condition

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Maxwell' s Equations

Case of a Linear Dielectric material :
no free charges, no free currents, nonmagnetic

$$\begin{cases} \nabla \times \mathcal{E} = -\dfrac{\partial B}{\partial t} & \nabla \cdot \mathcal{D} = 0 \\ \nabla \times H = \dfrac{\partial \mathcal{D}}{\partial t} & \nabla \cdot B = 0, \end{cases}$$

$$\mathcal{D} = \epsilon_0 \mathcal{E} + \mathcal{P} \qquad B = \mu_0 H$$

macroscopic Polarization = source terme $\vec{P} = \varepsilon_0 \underline{\underline{\chi}}^{(1)} \vec{E} + \vec{P}_{NL}$ **(Frequency domain)**

$$\vec{P}_{NL} = \varepsilon_0 \underline{\underline{\underline{\chi}}}^{(2)} \vec{E}\vec{E} + \varepsilon_0 \underline{\underline{\underline{\underline{\chi}}}}^{(3)} \vec{E}\vec{E}\vec{E} + \cdots$$ **(Frequency domain)**

Maxwell's Equation

Wave equation

In the time domain

$$\nabla \times \nabla \times \mathcal{E}(\boldsymbol{r}, t) + \quad \ldots$$

In the frequency domain

$$\nabla \times \nabla \times \boldsymbol{E}(\boldsymbol{r}, \omega) - \ldots$$

Maxwell's Equation

Wave equation

In the time domain

$$\nabla \times \nabla \times \boldsymbol{\mathcal{E}}(\boldsymbol{r},t) + \frac{1}{c^2}\frac{\partial^2 \boldsymbol{\mathcal{E}}(\boldsymbol{r},t)}{\partial t^2} = -\mu_0 \frac{\partial^2 \boldsymbol{\mathcal{P}}^{(1)}(\boldsymbol{r},t)}{\partial t^2} - \mu_0 \frac{\partial^2 \boldsymbol{\mathcal{P}}^{(NL)}(\boldsymbol{r},t)}{\partial t^2}$$

In the frequency domain

$$\nabla \times \nabla \times \boldsymbol{E}(\boldsymbol{r},\omega) - \frac{\omega^2}{c^2}\boldsymbol{E}(\boldsymbol{r},\omega) = \omega^2 \mu_0 \boldsymbol{P}^{(1)}(\boldsymbol{r},\omega) + \omega^2 \mu_0 \boldsymbol{P}^{(NL)}(\boldsymbol{r},\omega)$$

$\boldsymbol{P}^{(1)}(\boldsymbol{r},\omega) = \epsilon_0 \underline{\underline{\chi}}^{(1)}(\boldsymbol{r},\omega)\boldsymbol{E}(\boldsymbol{r},\omega)$ (local response)

$\underline{\underline{\epsilon}}(\boldsymbol{r},\omega) = 1 + \underline{\underline{\chi}}^{(1)}(\boldsymbol{r},\omega)$ (Relative permitivity)

$$\boxed{\nabla \times \nabla \times \boldsymbol{E}(\boldsymbol{r},\omega) = \frac{\omega^2}{c^2}\underline{\underline{\epsilon}}(\boldsymbol{r},\omega)\boldsymbol{E}(\boldsymbol{r},\omega) + \omega^2 \mu_0 \boldsymbol{P}^{(NL)}(\boldsymbol{r},\omega)}$$

Nonlinear Wave Equation in isotropic material

$$\nabla \times \nabla \times \boldsymbol{E}(\boldsymbol{r},\omega) = \nabla(\nabla \cdot \boldsymbol{E}) - \Delta \boldsymbol{E}$$

$\nabla \cdot (\underline{\underline{\epsilon}}\boldsymbol{E}) = 0$ Homogeneous and Isotropic Material : $\nabla \cdot \boldsymbol{E} = 0$

$$\Delta \boldsymbol{E}(\omega) + \frac{\omega^2}{c^2}\epsilon \boldsymbol{E}(\omega) = -\omega^2 \mu_0 \boldsymbol{P}_{NL}(\omega)$$

Considering the propagation of a plane wave along the direction z :

$$\boldsymbol{E}(z,\omega) = A(z)e^{\imath kz}\boldsymbol{e}$$

$$\frac{\partial^2 A(z)}{\partial z^2} + 2\imath k \frac{\partial A(z)}{\partial z} = -\frac{\omega^2}{\epsilon_0 c^2}\boldsymbol{e} \cdot \boldsymbol{P}_{NL}(z,\omega)e^{-\imath kz}$$

As $k^2(\omega) = \frac{\omega^2}{c^2}\epsilon(\omega)$ (dispersion relation)

Slowly varying amplitude approximation : $\left|\frac{\partial^2 A(z)}{\partial z^2}\right| \ll \left|2k\frac{\partial A(z)}{\partial z}\right|$

$$\boxed{\frac{\partial A(z)}{\partial z} = \frac{\imath\omega}{2\epsilon_0 n c}\boldsymbol{e} \cdot \boldsymbol{P}_{NL}(z,\omega)e^{-\imath kz}}$$ ★★★

Conclusion

- Nonlinear wave equation :

$$P_{NL}(z,\omega) = \Pi_{NL}(z,\omega)\, e^{\imath k_p(\omega)\cdot r}$$

$$\frac{\partial A(z)}{\partial z} = \frac{\imath\omega}{2\epsilon_0 nc} e \cdot P_{NL}(z,\omega) e^{-\imath kz}$$

$$\frac{\partial A(z)}{\partial z} = \frac{\imath\omega}{2\epsilon_0 nc} e \cdot \Pi_{NL}(z,\omega) e^{-\imath\Delta kz}$$

- **Efficient Energy transfer requires :**

  - Non-zero Nonlinear Polarization amplitude @ ω

  - Non-zero projection between the electric field and the NL polarization $\vec{e}\,.\,\vec{P}_{NL} \neq 0$

  - phase matching condition ***Δk=0***

NONLINEAR OPTICS

Nonlinear wave equation: application to SHG
& Phase matching condition

- Maxwell's equations

- Nonlinear wave equation in a isotropic material

$$\frac{\partial A(z)}{\partial z} = \frac{\imath\omega}{2\epsilon_0 nc} e \cdot P_{NL}(z,\omega) e^{-\imath kz}$$

➢ **Application : Second Harmonic Generation (SHG)**

=> SEE TUTORIAL n° 1

➢ Discussion about the phase matching condition

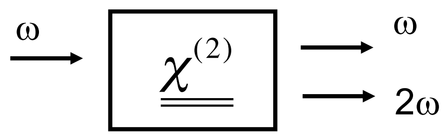
- Propagation in a linear anisotropic material

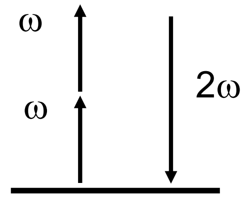
- Stationary nonlinear wave equation

- Phase Matching considerations

2nd Harmonic Generation

Assumption : Lossless medium





1. Undepleted pump approximation regime

$A_\omega(z)$=Cste

$$\begin{cases} \dfrac{dA_\omega(z)}{dz} = 0 \\ \dfrac{dA_{2\omega}(z)}{dz} = \dfrac{\imath(2\omega)}{2\epsilon_0 n_{2\omega} c} \boldsymbol{e}_{2\omega} \cdot \boldsymbol{P}_{NL}(z, 2\omega) e^{-\imath \boldsymbol{k}_{2\omega}\cdot \boldsymbol{z}} \end{cases}$$

$$\boldsymbol{P}_{NL}(z, 2\omega) = \epsilon_0 \underline{\underline{\chi}}^{(2)}(2\omega;\omega,\omega)\boldsymbol{E}(z,\omega)\boldsymbol{E}(z,\omega)$$
$$= \epsilon_0 \underline{\underline{\chi}}^{(2)}(2\omega;\omega,\omega)\boldsymbol{e}_\omega \boldsymbol{e}_\omega A_\omega^2(z) e^{\imath 2\boldsymbol{k}_\omega \cdot \boldsymbol{z}}$$

$$\Rightarrow \quad \boxed{\frac{dA_{2\omega}(z)}{dz} = \frac{\imath(2\omega)}{2n_{2\omega}c}\chi_{\text{eff}}^{(2)} \; A_\omega^2(z) e^{\imath \Delta \boldsymbol{k}\cdot \boldsymbol{z}}}$$

Wavevector missmatch:

$$\Delta \boldsymbol{k} = 2\boldsymbol{k}_\omega - \boldsymbol{k}_{2\omega}$$

Effective nonlinear susceptibility

$$\chi_{\text{eff}}^{(2)} = \boldsymbol{e}_{2\omega} \cdot \underline{\underline{\chi}}^{(2)}(2\omega;\omega,\omega)\boldsymbol{e}_\omega \boldsymbol{e}_\omega$$

2nd Harmonic Generation

1. Undepleted pump approximation regime

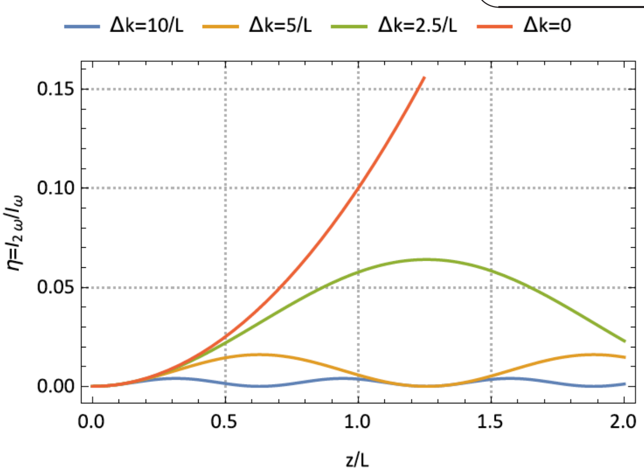
Solution : Intensity evolution

$$I_{2\omega}(z) = \frac{(2\omega)^2}{2\epsilon_0 n_\omega^2 n_{2\omega} c^3} \left|\chi_{\text{eff}}^{(2)}\right|^2 \sin^2\left(\frac{\Delta k}{2} z\right) \frac{I_\omega^2}{(\Delta k)^2}$$
$$= \frac{(2\omega)^2}{8\epsilon_0 n_\omega^2 n_{2\omega} c^3} \left|\chi_{\text{eff}}^{(2)}\right|^2 \; \text{sinc}^2(\Delta k z/2) \; I_\omega^2 \; z^2.$$

Field intensity :
$I = 2nc\epsilon_0|E_0|^2,$

SHG efficiency :

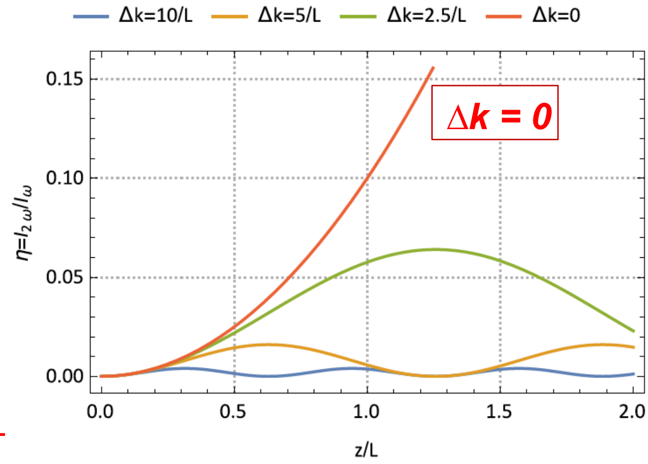
$$\eta_{\text{SHG}} = \frac{I_{2\omega}}{I_\omega} = \frac{(2\omega)^2}{8\epsilon_0 n_\omega^2 n_{2\omega} c^3} \left|\chi_{\text{eff}}^{(2)}\right|^2 \; \text{sinc}^2(\Delta k z/2) \; I_\omega \; z^2.$$



2nd Harmonic Generation

1. Undepleted pump approximation regime

SHG efficiency :

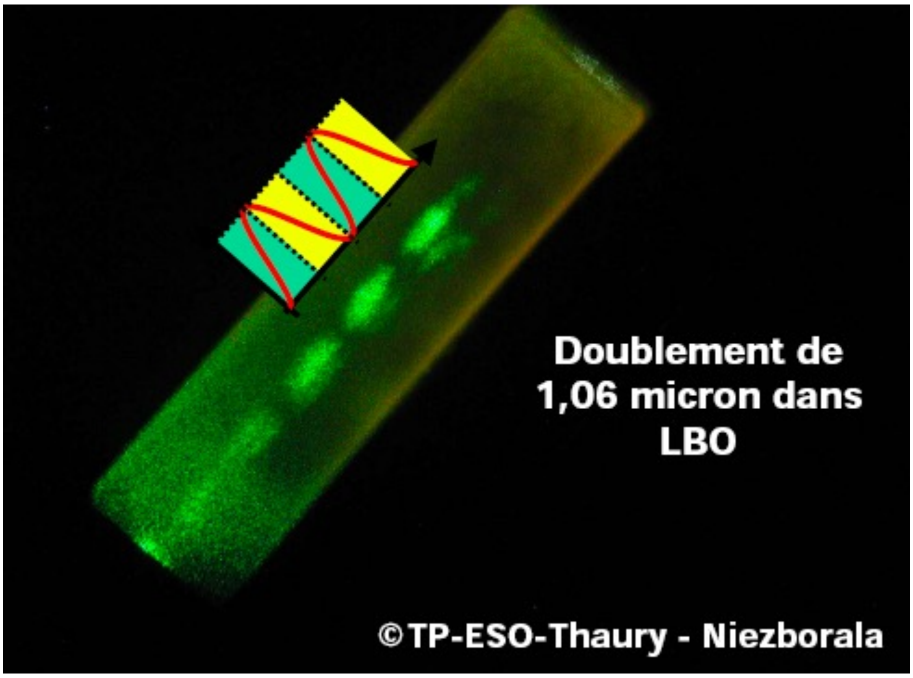


Conclusions :

- Non-phasematched situation : generation @ 2ω occurs on a typical length $L_{coh}=\pi/\Delta k$, called ***coherent buildup length (coherence length)***
- Intensity @ 2ω is proportional to $I_\omega^2/\Delta k^2$
- The conversion efficiency $I_{2\omega}/I_\omega$ is proportional to I_ω *(need to focus the beam @ ω to increase the efficiency)*
- ***Efficiency Max. :*** *requires to fulfill the* ***phasematching condition: $\Delta k=0$***

2 - 2nd Harmonic Generation

Non-phasematched situation : ***Δk≠0***



NONLINEAR OPTICS

Nonlinear wave equation: application to SHG & Phase matching condition

- Maxwell's equations
- Nonlinear wave equation in a isotropic material
  - ➢ Application : Second Harmonic Generation (SHG)
  - ➢ **Discussion about the phase matching condition**
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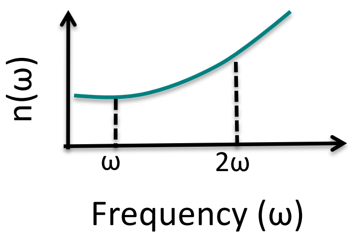
Phase matching condition

About the difficulty to achieve the phase matching condition :

In general, the refractive index for lossless materials shows a NORMAL DISPERSION : the refractive index is increasing with the frequency



Case of 2nd Harm. Gen. :

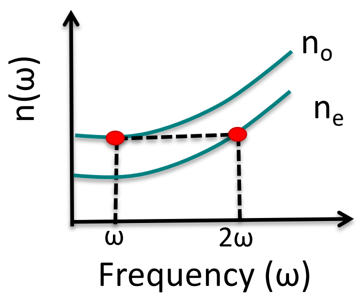
Phase matching condition implies :

n(ω)=n(2ω)

IMPOSSIBLE !

The most common procedure for achieving the phase matching condition :

Use of birefringence properties of crystals.



Phase matching condition

Nonlinear wave equation : $$\frac{\partial A(z)}{\partial z} = \frac{\imath\omega}{2\epsilon_0 nc} e \cdot P_{NL}(z,\omega) e^{-\imath kz}$$

Substituting $P_{NL}(z,\omega) = \Pi_{NL}(z,\omega)\, e^{\imath \boldsymbol{k}_p(\omega)\cdot \boldsymbol{r}}$
in the nonlinear wave equation :

$$\frac{\partial A(z)}{\partial z} = \frac{\imath\omega}{2\epsilon_0 nc} e \cdot \Pi_{NL}(z,\omega) e^{-\imath \Delta kz}$$

The phase miss-match term $\Delta kz = (k_p - k)\cdot z$

Assumption : weak nonlinear interaction (or parametric approx)

$\Pi_{NL}(z,\omega) \simeq$ Const. along z

Solution of the wave equation :

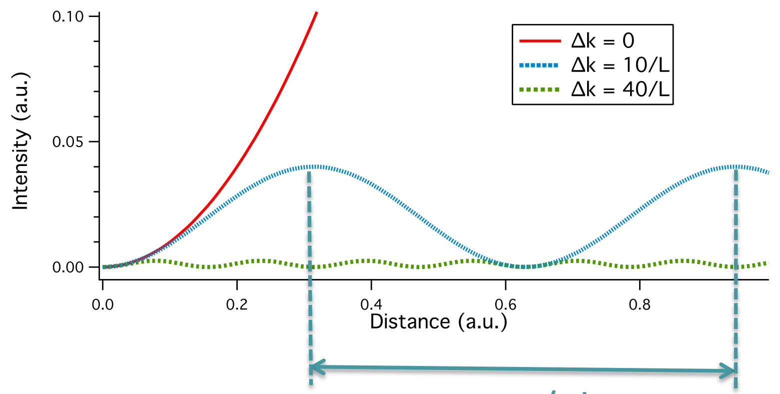
$$I(z) = \frac{\omega^2}{2nc\epsilon_0} |e \cdot \Pi_{NL}(\omega)|^2 \operatorname{sinc}^2\left(\frac{\Delta kL}{2}\right) L^2$$

Phase matching condition

Nonlinear wave equation : $$\frac{\partial A(z)}{\partial z} = \frac{\imath\omega}{2\epsilon_0 nc} e \cdot P_{NL}(z,\omega) e^{-\imath kz}$$

Solution of the wave equation : under a weak nonlinear interaction

$$I(z) = \frac{\omega^2}{2nc\epsilon_0} |e \cdot \Pi_{NL}(\omega)|^2 \operatorname{sinc}^2\left(\frac{\Delta kL}{2}\right) L^2$$



$L_c = \pi/\Delta k$ ***: coherence length***

When *Δk≠0*, sinusoïdal evolution of *I(z)*
When *Δk=0*, I(z) grows as z^2 (under the undepleted-pump approximation)

A physical picture : Free and driven waves - Phase Matching Condition

Nonlinear wave equation $$\frac{d^2E(\omega)}{dz^2} + k^2E(\omega) = -\omega^2\mu_0\Pi_{NL}(\omega)\,e^{\imath k_p z}$$

Complete solution $E = E_o(z) + E_f(z)$

With $E_o(z)$ solution of $\frac{d^2E(\omega)}{dz^2} + k^2\epsilon E(\omega) = 0$ **= FREE running WAVE**

and $E_f(z)$ driven solution of the wave equation **= DRIVEN WAVE**

- **Sets of solution : general form**

$$E_o(z) = eA_0e^{\imath kz}$$
$$E_f(z) = eA_fe^{\imath k_p z}$$

With $A_f \simeq$ Const. in undepleted wave approximation, considering $P_{NL}(\omega)$ independant of *z*

Free and Driven Waves...

- Solution for the driven wave

$$A_f \simeq \frac{\omega}{2\epsilon_0 nc\Delta k}e.\Pi_{NL}$$

- Complete solution $$E = e\left[A_0e^{\imath kz} + A_fe^{\imath k_p z}\right]$$

Free wave + driven wave

- boundary condition $E(z=0) = 0e \Rightarrow A_f = -A_o$

Intensity evolution $$I(z) = \frac{\omega^2}{2nc\epsilon_0}|e\cdot\Pi_{NL}(\omega)|^2\,\mathrm{sinc}^2\left(\frac{\Delta kL}{2}\right)L^2$$

INTERPRETATION

When *Δk≠0*, sinusoïdal evolution of *I(z)* : successive constructive and destructive interference between the free wave and the driven wave (induced by P_{NL})
When *Δk=0*, constructive interference and I(z) behaves as z^2 (as long as the undepleted-pump approximation is valid)

Student activities

To complete, read the lecture notes :
➜ sections 3.2 and 3.3

Refresher about the birefringence properties of anisotropic materials