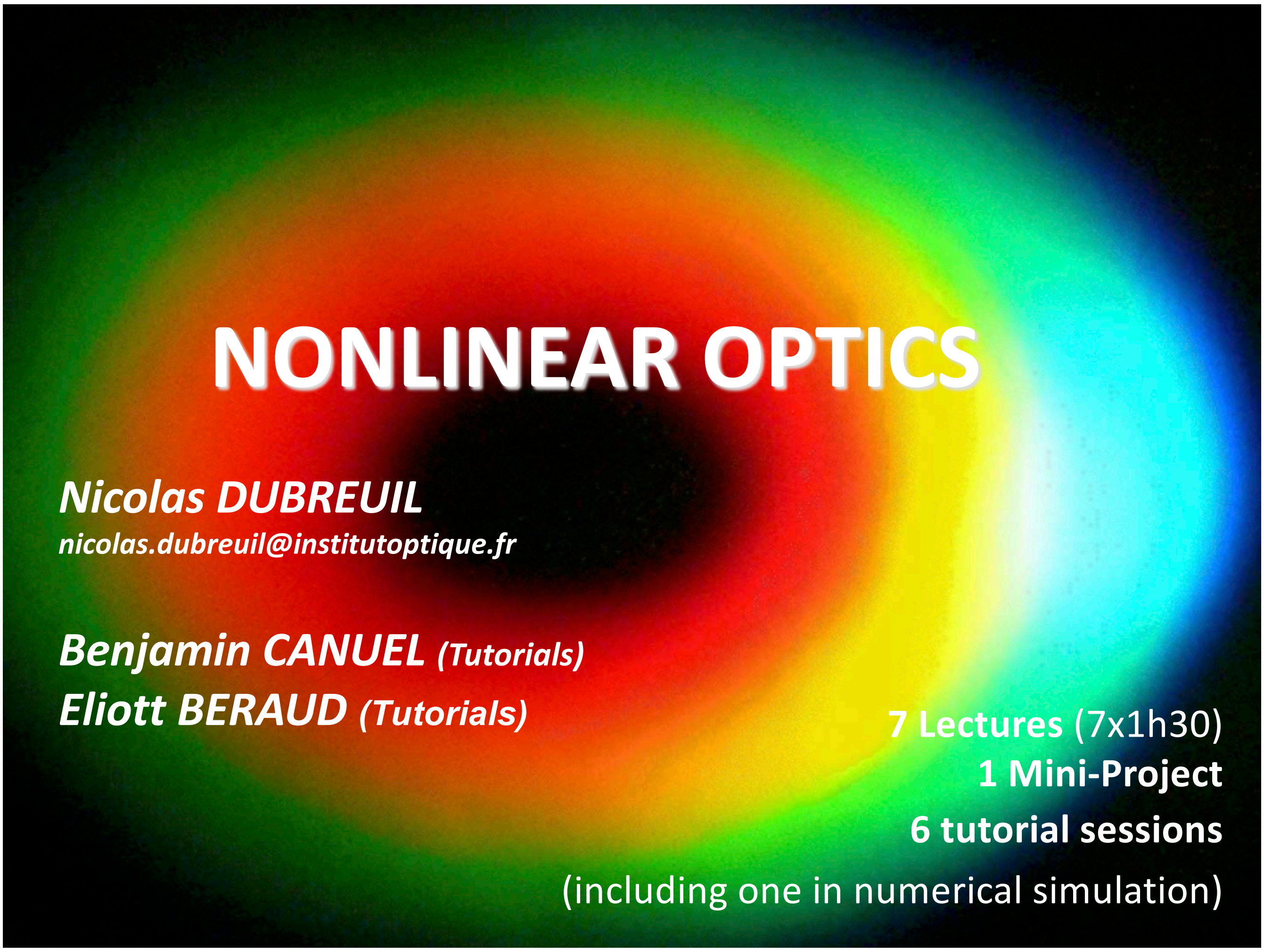


Optics in nonlinear regime ?

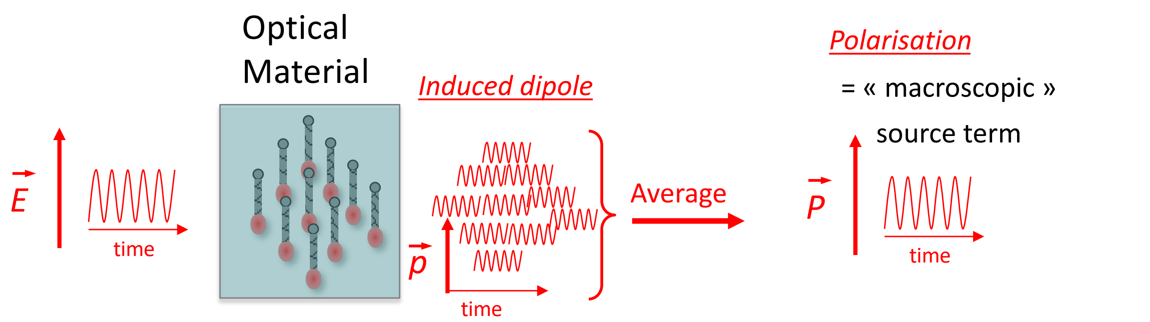
- **Introduction to OPTICS in NONLINEAR REGIME**
    - **Which applications ?**
        - **Physical origin of the nonlinearities ?**

Optics in nonlinear regime ?

- **Introduction to OPTICS in NONLINEAR REGIME**
  - Which applications ?
    - Physical origin of the nonlinearities ?

Introduction to Nonlinear Optics

- **Response of a material subject to an incident EM wave at ω**



$E(t) = A\cos(\omega t - kz)$ —LINEAR REGIME→ $P(t) \propto R^{(1)}\cos(\omega t - kz)$

LINEAR RESPONSE of the material

WHAT HAPPENS WHEN increasing the MAGNITUDE of electric field amplitude ?

Introduction to Nonlinear Optics

- **Case of a NONLINEAR MEDIUM :**

The nonlinear response of the medium can be expressed as

$$P(t) = \varepsilon_0\chi^{(1)}E(t)+\varepsilon_0\chi^{(2)}E(t)E(t)+\varepsilon_0\chi^{(3)}E(t)E(t)E(t)$$

$$= \underbrace{\varepsilon_0\chi^{(1)}E(t)}_{\text{Linear response}}+\underbrace{\varepsilon_0\chi^{(2)}E^2(t)+\varepsilon_0\chi^{(3)}E^3(t)}_{\text{2nd Order + 3rd Order Nonlinear Response}}+\cdots$$

(we have assumed that the medium have an instantaneous response - Case of a lossless and a dispersionless medium) :

IMPORTANT COMMENT : Nonlinear interactions are governed by the <u>magnitude of the electric fields</u>

Introduction to Nonlinear Optics

- **Case of a NONLINEAR MEDIUM**

The nonlinear response of the medium can be expressed as

$$P(t) = \varepsilon_0\chi^{(1)}E(t)+\varepsilon_0\chi^{(2)}E(t)E(t)+\varepsilon_0\chi^{(3)}E(t)E(t)E(t)$$

$$= \underbrace{\varepsilon_0\chi^{(1)}E(t)}_{\text{Linear response}}+\underbrace{\varepsilon_0\chi^{(2)}E^2(t)+\varepsilon_0\chi^{(3)}E^3(t)}_{\text{2nd Order + 3rd Order Nonlinear Response}}+\cdots$$

Considering a z propagative EM wave @ ω

$$E(t) = A\cos(\omega t+kz)$$

1st Order

$$P(t)\propto\chi^{(1)}A\cos(\omega t+kz)$$

Source term @ ω

2nd Order

$$P(t)\propto\chi^{(2)}A^2\cos^2(\omega t+kz)^2$$

$$\propto\chi^{(2)}A^2\cos(2\omega t+2kz)+\chi^{(2)}A^2$$

Source term @ 2ω **!!!**

Introduction to Nonlinear Optics

- **Case of a NONLINEAR MEDIUM**

The nonlinear response of the medium can be expressed as :

$$P(t) = \varepsilon_0\chi^{(1)}E(t) + \varepsilon_0\chi^{(2)}E(t)E(t) + \varepsilon_0\chi^{(3)}E(t)E(t)E(t)$$

$$= \varepsilon_0\chi^{(1)}E(t) + \varepsilon_0\chi^{(2)}E^2(t) + \varepsilon_0\chi^{(3)}E^3(t) + \cdots$$

Linear response | 2nd Order + 3rd Order Nonlinear Response

Considering a z propagative EM wave @ ω

1st Order

$$P(t) \propto \chi^{(1)}A\cos(\omega t + kz)$$

Source term @ ω

3rd Order

$$E(t) = A\cos(\omega t + kz)$$

$$P_{NL}(t) \propto \chi^{(3)}A^3\cos^3(\omega t + kz)$$

$$\propto \chi^{(3)}A^3\cos(3\omega t + 3kz) + \chi^{(3)}A^2A\cos(\omega t + kz) + \cdots$$

Source term @ 3ω
Third harmonic generation

Source term @ ω
Optical Kerr effect

Example : Optical Kerr Effect

✓ **Variation of the refractive index**

$$P(t) = \varepsilon_0\chi^{(1)}E(t) + \varepsilon_0\chi^{(3)}E^3(t)$$

Case of a lossless medium

$$P(t) = P_L(t) + P_{NL}(t) \propto \chi^{(1)}A\cos(\omega t + kz) + \chi^{(3)}A^2A\cos(\omega t + kz)$$

$$P(t) \propto \left(\chi^{(1)} + \chi^{(3)}A^2\right)A\cos(\omega t + kz)$$ NONLINEAR Regime

$$P_L(t) \propto \chi^{(1)} A\cos(\omega t + kz)$$ LINEAR Regime

Directly related to the refractive index

Kerr effect induces a variation of the refractive index directly proportional to the wave intensity.

Considering a *z* propagative EM wave @ ω

$$E(t) = A\cos(\omega t + kz)$$

$$I \propto A^2$$ **Wave intensity**

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 : $\boldsymbol{E}(\omega) = \left| \begin{array}{l} E_i(\omega) \\ E_j(\omega) \\ E_k(\omega) \end{array} \right.$

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

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 :

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

Optics in nonlinear regime ?

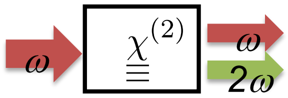
- Introduction to OPTICS in NONLINEAR REGIME

- **Which applications ?**

- Physical origin of the nonlinearities ?

2nd order nonlinear interactions

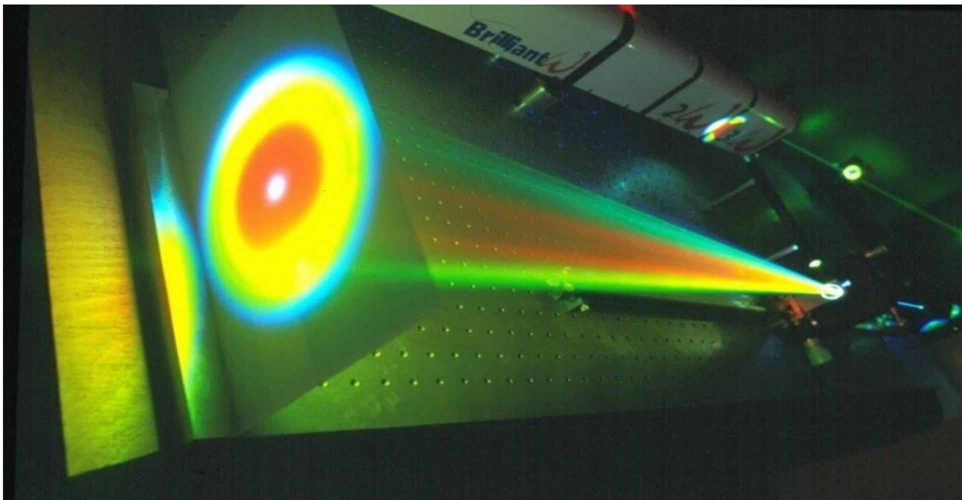
Second Harmonic Generation



Application: Green Laser Pointer

Optical Parametric Fluorescence & Amplification

Optical source with a wide frequency tunability

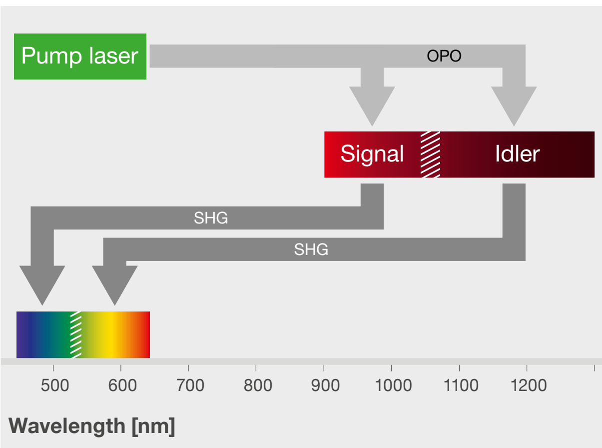


Credit Photo: Institut d'Optique

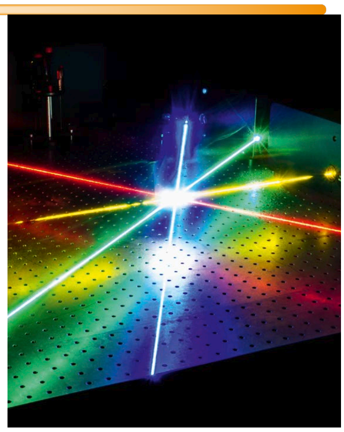
2nd order nonlinear interactions

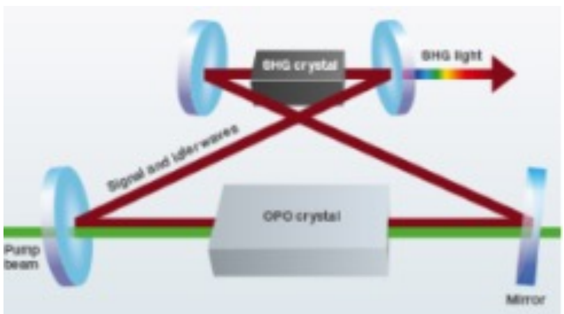
Optical Parametric Oscillator

Optical source with broad wavelength tunability



HÜBNER





https://www.hubner-photonics.com

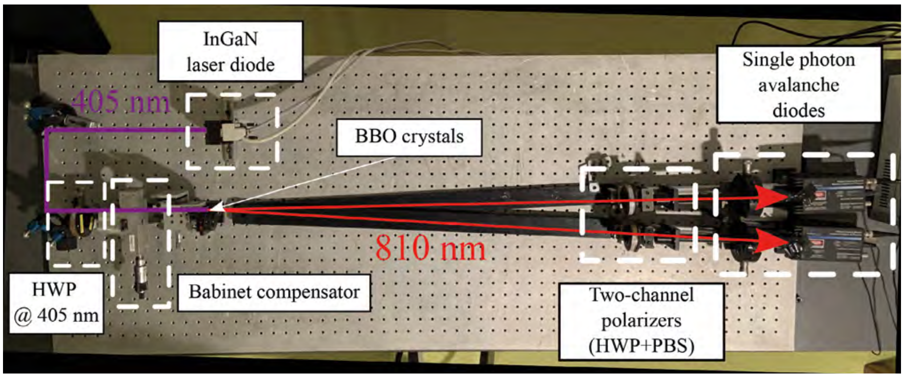
Credit Photo: Institut d'Optique

2^{nd} order nonlinear interactions

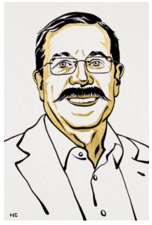
Optical Parametric Fluoresence effect

Source of polarization entangled state pairs of photons

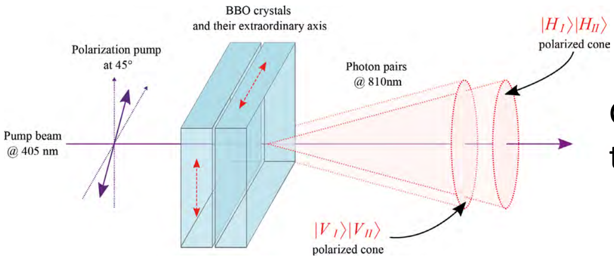
Quantum Optics



Experiment to test the violation of the Bell's inequalities (Labwork @ IOGS)



Alain Aspect
Nobel Prize in Physics 2022

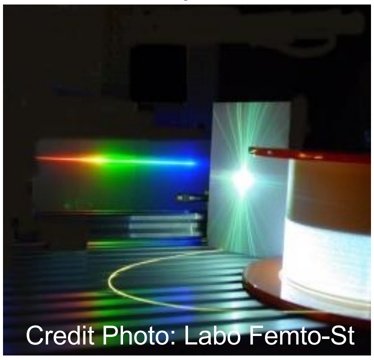


Generation of photon pairs in the superposition state :

$$|\psi_{2ph}\rangle = \frac{1}{\sqrt{2}}\left(|V_I\rangle|V_{II}\rangle + e^{i\varphi_0}|H_I\rangle|H_{II}\rangle\right)$$

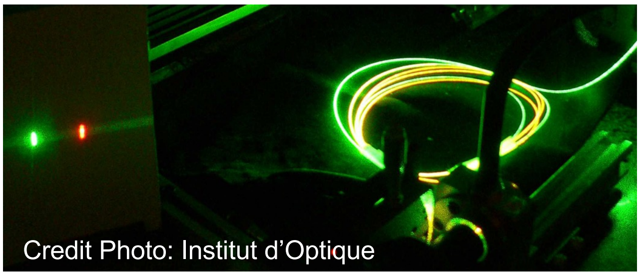
3^{rd} order nonlinear interactions

Supercontinuum Generation in nonlinear optical fibers

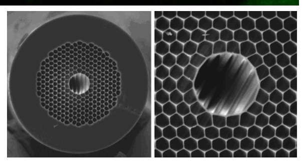


Credit Photo: Labo Femto-St

Raman scattering in a hollow-core photonic crystal fibre filled with liquid or gas



Credit Photo: Institut d'Optique

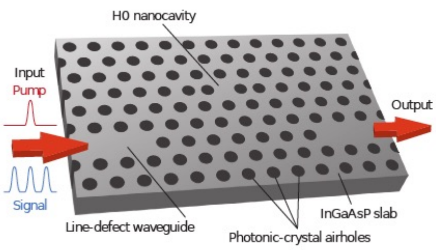


Hollow-core fibre

Optical Kerr Effect

Refractive index variation α Optical Intensity

All-optical swithing in a µcavity with pump energy of 10 fJ !



Raman scattering

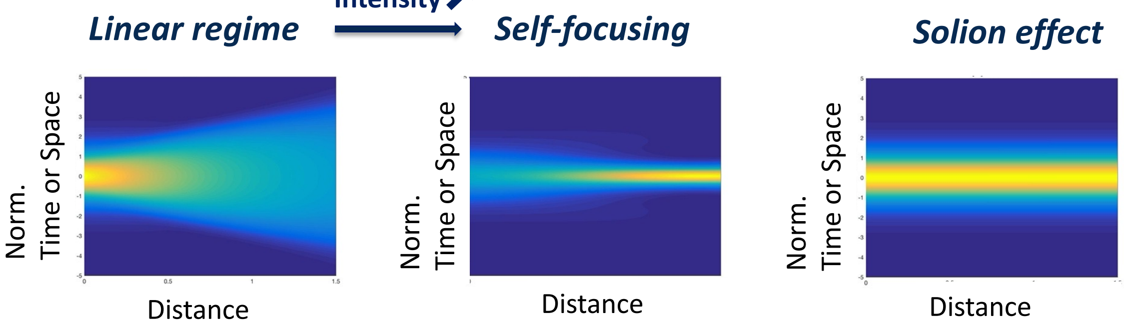
3rd order nonlinear interactions

Optical solitons

propagation of a wave packet (pulse or spatial beam) through a pure Kerr medium ⇒ ***refractive index variation proportional to the wave intensity.***

⇒ Optical Kerr lens effect (in time or space) !!

Intensity ↗



propagation of the wave packet WITHOUT any deformation

3rd order nonlinear interactions

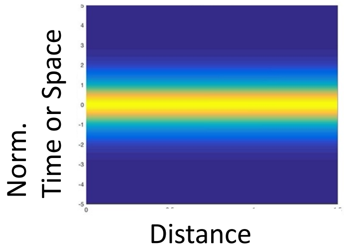
Optical solitons

propagation of a wave packet (pulse or spatial beam) through a pure Kerr medium ⇒ ***refractive index variation proportional to the wave intensity.***

⇒ Optical Kerr lens effect (in time or space) !!

Mascaret waves on the Gironde estuary

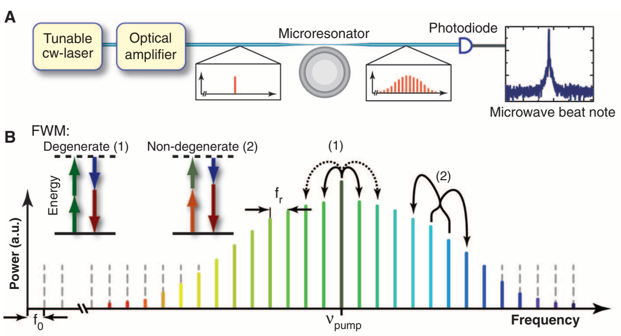
Solion effect



propagation of the wave packet WITHOUT any deformation

3rd order nonlinear interactions

Generation of optical Frequency Combs in nonlinear micro-cavities



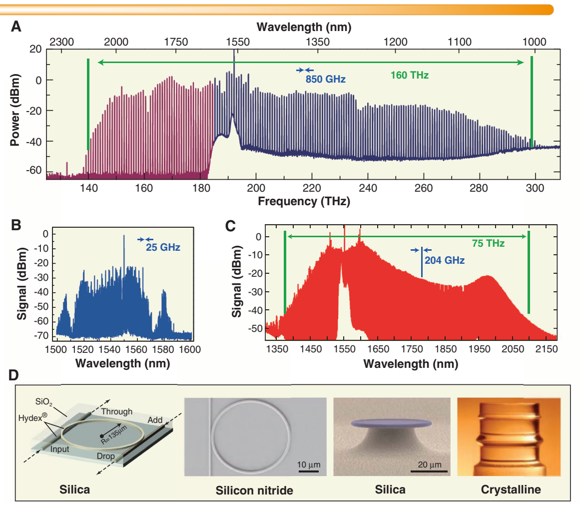
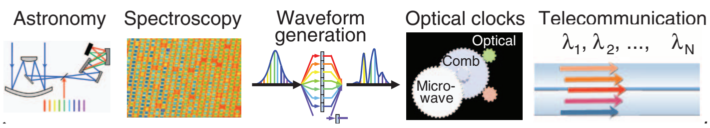


Fig. 3. Microresonator-based frequency combs. **(A)** Spectrum of an octave-spanning frequency comb generated using a silica microtoroidal resonator *(24)*. **(B)** An optical frequency comb generated using a crystalline CaF_2 resonator with a mode spacing of 25 GHz *(27)*. **(C)** Optical spectrum covering two-thirds of an octave (with a mode spacing of 204 GHz) generated using an integrated SiN resonator *(31)*. **(D)** Experimental systems in which frequency combs have been generated (from left to right): Silica waveguides on a chip (Hydex glass) *(32)*, chip-based silicon nitride (SiN) ring resonators *(30)* and waveguides, ultrahigh Q toroidal microresonators *(24)* on a silicon chip, and ultrahigh *Q* millimeter-scale crystalline resonators *(27)*.

Applications :



From Kippenberg, Science (2011)

INSTITUT d'OPTIQUE GRADUATE SCHOOL

Optics in nonlinear regime ?

- Introduction to OPTICS in NONLINEAR REGIME
  - Which applications ?
    - **Physical origin of the nonlinearities ?**

Nonlinear Optics

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

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

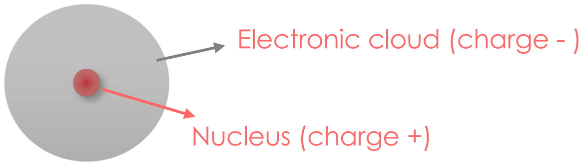
  - **Origin of the nonlinearities : classical anharmonic oscillator (classical model)**

Induced dipole and macroscopic polarization

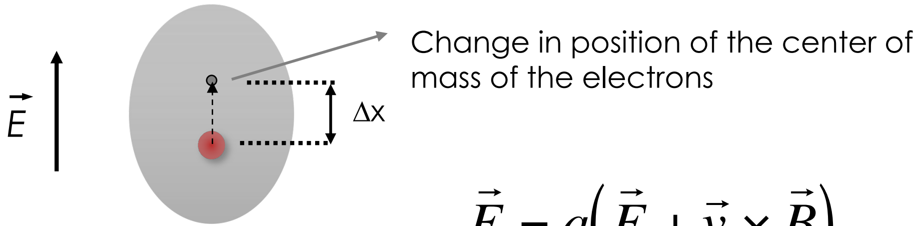
✓ Microscopic scale

A simplistic description of the interaction between a wave and an atom :

- Simplistic model for an atom =



- An applied static electric field acts on the electrons trajectories



$$\vec{F} = q\left(\vec{E} + \vec{v} \times \vec{B}\right)$$

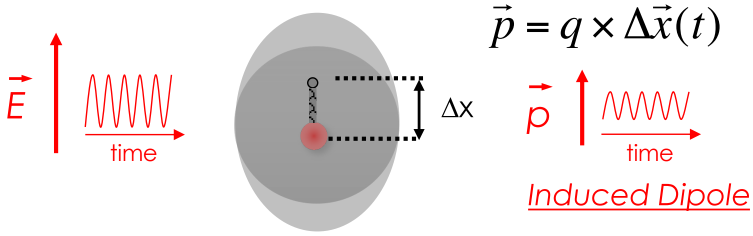
Lorentz Force

Induced dipole and macroscopic polarization

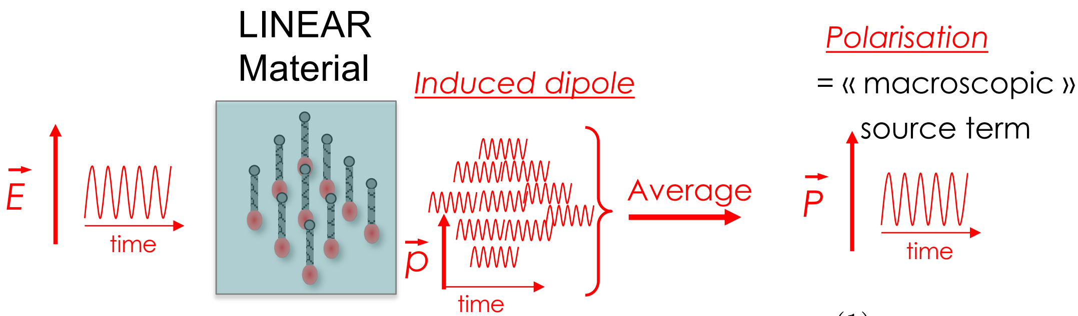
The magnetic part of the Lorentz force is negligible

- **With an applied EM fiels = temporal deformation of the electronic cloud**

$$\vec{p} = q \times \Delta\vec{x}(t)$$



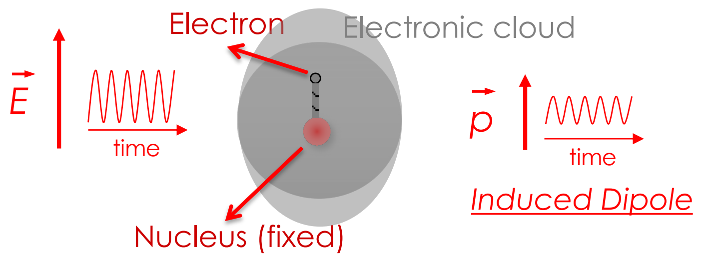
- **LINEAR MEDIUM case made of N atoms (N identical dipoles)**



$E(t) = A\cos(\omega t - kz)$ → $P(t) \propto R^{(1)}\cos(\omega t - kz)$

Classical harmonic oscillator model

- ***Classical anharmonic oscillator - Description***



Induced dipole (microscopic quantity) : $\boldsymbol{p}(z,t) = -e\,x(z,t)\boldsymbol{x}$

Polarization (MACROscopic quantity) : $\boldsymbol{\mathcal{P}}(z,t) = N\boldsymbol{p}(z,t)$

} *x(z,t) ??*

- ***Equation of motion***

$$\frac{d^2x}{dt^2} + \alpha\frac{dx}{dt} + \omega_0^2 x = \frac{-e}{m}\boldsymbol{x}\cdot\boldsymbol{\mathcal{E}}(z,t)$$

Damping term — Restoring force — Driven Coulomb force

Linear polarization

Harmonic oscillator : equation of motion

$$\frac{d^2x}{dt^2} + \alpha\frac{dx}{dt} + \omega_0^2 x = \frac{-e}{m}\left[A(\omega)e^{-\imath(\omega t - kz)} + A(-\omega)e^{+\imath(\omega t - kz)}\right]\boldsymbol{x}\cdot\boldsymbol{x}.$$

....

Driven solution $x^{(1)}(z,t) = a(\omega)e^{-\imath(\omega t - kz)} + a(-\omega)e^{+\imath(\omega t - kz)}$

Induced dipole $\boldsymbol{p}^{(1)}(z,t) = \alpha^{(1)}(\omega)A(\omega)e^{-\imath(\omega t - kz)}\boldsymbol{x} + CC.$

Linear polarizability (microscopic quantity) $\alpha^{(1)} = \frac{e^2}{m\mathcal{D}(\omega)}$ $\mathcal{D}(\omega) = \omega_0^2 - \omega^2 - \imath\alpha\omega$

Macroscopic Polarization

Linear susceptibility

$$\boldsymbol{\mathcal{P}}^{(1)}(z,t) = \epsilon_0\chi^{(1)}(\omega)\boldsymbol{E}(\omega)e^{-\imath\omega t} + CC.$$

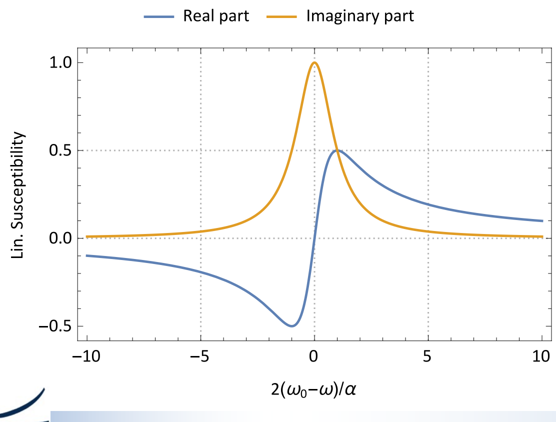
Linear polarization

Harmonic oscillator : equation of motion

$$\frac{d^2x}{dt^2} + \alpha\frac{dx}{dt} + \omega_0^2 x = \frac{-e}{m}\left[A(\omega)e^{-\imath(\omega t - kz)} + A(-\omega)e^{+\imath(\omega t - kz)}\right]\boldsymbol{x}\cdot\boldsymbol{x}.$$

$$\boldsymbol{\mathcal{P}}^{(1)}(z,t) = \epsilon_0\chi^{(1)}(\omega)\boldsymbol{E}(\omega)e^{-\imath\omega t} + CC.$$

Linear Susceptibility



$$\chi^{(1)}(\omega) = \frac{Ne^2}{\epsilon_0 m(\omega_0^2 - \omega^2 - \imath\alpha\omega)}$$

$$\chi^{(1)}(\omega) = \chi'(\omega) + \imath\chi''(\omega)$$

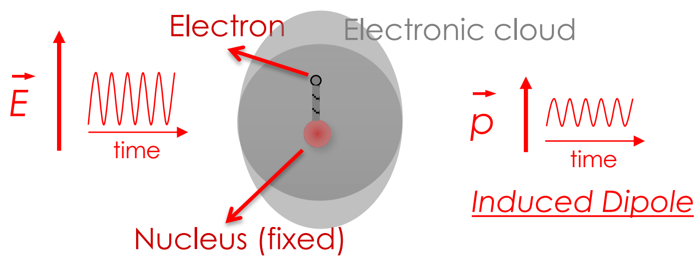
Dispersion

Absorption (or amplification)

Lorentzian line shape

Classical anharmonic oscillator model

- ***Classical anharmonic oscillator - Description***



Induced dipole (microscopic quantity) : $\boldsymbol{p}(z,t) = -e\,x(z,t)\boldsymbol{x}$

Polarization (MACROscopic quantity) : $\boldsymbol{\mathcal{P}}(z,t) = N\boldsymbol{p}(z,t)$

x(z,t) ??

- ***Equation of motion***

$$\frac{d^2x}{dt^2} + \alpha\frac{dx}{dt} + \omega_0^2 x + \beta x^2 + \gamma x^3 + \cdots = \frac{-e}{m}\boldsymbol{x}\cdot\boldsymbol{\mathcal{E}}(z,t)$$

Classical anharmonic oscillator model

- ***Equation of motion***

$$\frac{d^2x}{dt^2} + \alpha\frac{dx}{dt} + \omega_0^2 x + \beta x^2 + \gamma x^3 + \cdots = \frac{-e}{m} x \cdot \boldsymbol{\mathcal{E}}(z,t)$$

Damping term — Restoring force — Driven Coulomb force

Solution : perturbation method, taking into account $\omega_0^2 x \gg \beta x^2 \gg \gamma x^3$

$$x = \lambda x^{(1)} + \lambda^2 x^{(2)} + \lambda^3 x^{(3)} + \ldots$$

$$x^{(1)} \gg x^{(2)} \gg x^{(3)}$$

2nd Order Nonlinear Polarization

Anharmonic oscillator : equation of motion

$$\frac{d^2x}{dt^2} + \alpha\frac{dx}{dt} + \omega_0^2 x + \beta x^2 = \frac{-e}{m} x \cdot \boldsymbol{\mathcal{E}}(z,t)$$

Driven solution $x(z,t) = \lambda x^{(1)}(z,t) + \lambda^2 x^{(2)}(z,t) + \cdots$

$$\frac{d^2x^{(2)}}{dt^2} + \alpha\frac{dx^{(2)}}{dt} + \omega_0^2 x^{(2)} = -\beta\left(x^{(1)}\right)^2$$

$$x^{(2)}(z,t) = b(0) + b(2\omega)e^{-2i(\omega t - kz)} + b(-2\omega)e^{+2i(\omega t - kz)}$$

$$\begin{cases} b(0) = \dfrac{-2\beta e^2 |A|^2(\omega)}{m^2 \mathcal{D}(0)\mathcal{D}(\omega)\mathcal{D}(-\omega)} \\ b(\pm 2\omega) = \dfrac{-\beta e^2 A^2(\pm\omega)}{m^2 \mathcal{D}(\pm 2\omega)\mathcal{D}(\pm\omega)\mathcal{D}(\pm\omega)} \end{cases}$$

With : $D(\omega) = \omega_0^2 - \omega^2 - i\alpha\omega$

2nd Order Nonlinear Polarization

Anharmonic oscillator : equation of motion

$$\mathcal{P}(z,t) = \mathcal{P}^{(1)}(z,t) + \mathcal{P}^{(2)}(z,t)$$
$$= P^{(2)}(0) + P^{(1)}(\omega)e^{-\imath\omega t} + P^{(2)}(2\omega)e^{-2\imath\omega t} + CC.$$

$$P^{(2)}(0) = 2\epsilon_0\chi^{(2)}(\omega,-\omega)E(\omega)E(-\omega)\boldsymbol{x}$$

Optical RECTIFICATION
(induces static electric field)

$$P^{(2)}(2\omega) = \epsilon_0\chi^{(2)}(\omega,\omega)E(\omega)E(\omega)\boldsymbol{x}$$

2nd Harmonic Generation
creation of an electric field with a 2ω frequency component

2nd order Nonlinear Susceptibility $\chi^{(2)}(\omega_1,\omega_2) = \dfrac{N\alpha^{(2)}(\omega_1,\omega_2)}{\epsilon_0}$

$$\alpha^{(2)}(\omega_1,\omega_2) = \frac{\beta e^3}{m^2\mathcal{D}(\omega_1+\omega_2)\mathcal{D}(\omega_1)\mathcal{D}(\omega_2)}$$

2nd Order Nonlinear Polarization

Anharmonic oscillator : equation of motion

Conclusion & Comments

• The macroscopic polarization induced inside the material is then given by the sum :

$$\boldsymbol{P}(z,t) = \boldsymbol{P}^{(1)}(z,t) + \boldsymbol{P}^{(2)}(z,t)$$
$$= \boldsymbol{P}^{(2)}(0) + \boldsymbol{P}^{(1)}(\omega)e^{-\imath(\omega t-kz)} + \boldsymbol{P}^{(2)}(2\omega)e^{-2\imath(\omega t-kz)} + CC.$$

With : $\boldsymbol{P}^{(2)}(0) = 2\epsilon_0\chi^{(2)}(\omega,-\omega)E(\omega)E(-\omega)\boldsymbol{x}$

$\boldsymbol{P}^{(2)}(2\omega) = \epsilon_0\chi^{(2)}(\omega,\omega)E(\omega)E(\omega)\boldsymbol{x}$ (Complex amplitudes)

$$\chi^{(2)}(\omega_3;\omega_1,\omega_2) = \frac{N\,e^3\beta}{\epsilon_0 m^2 D(\omega_3)D(\omega_1)D(\omega_2)}$$

With : $\omega_3 = \omega_1 + \omega_2$

$D(\omega) = \omega_0^2 - \omega^2 - i\alpha\omega$

• For **non-resonant interactions** ($\omega \ll \omega_0$) ⇒ $\chi^{(2)}$ **is real and independent of the frequency**

• On the other hand, strong enhancement of the nonlinear susceptibility is expected once ω or 2ω (or both) is close to a material transition (@ω_0) (but with detrimental additional absorption)

• Phase mismatching between the polarization component @ 2ω and the free propagative wave @ 2ω ⇒ wavevector related to P(2ω) ≠ wavevector of E(2ω)

2k(ω) ≠ k(2ω)

3rd Order Nonlinear Polarization

Anharmonic oscillator : equation of motion

Case of a centro-symmetric material

$$\frac{d^2x}{dt^2} + \alpha\frac{dx}{dt} + \omega_0^2 x + \gamma x^3 = \frac{-e}{m} x \cdot \boldsymbol{\mathcal{E}}(z,t)$$

Driven solution

$$x(z,t) = \lambda x^{(1)}(z,t) + \lambda^2 x^{(2)}(z,t) + \lambda^3 x^{(3)}(z,t) \Rightarrow x^{(2)} = 0$$

Linear and Nonlinear Polarization

$$\begin{aligned}\boldsymbol{\mathcal{P}}(z,t) &= \boldsymbol{\mathcal{P}}^{(1)}(z,t) + \boldsymbol{\mathcal{P}}^{(3)}(z,t)\\ &= \boldsymbol{P}(\omega)e^{-\imath\omega t} + \boldsymbol{P}(3\omega)e^{-3\imath\omega t} + CC\end{aligned}$$

$$\boldsymbol{P}^{(3)}(3\omega) = \epsilon_0 \chi^{(3)}(\omega,\omega,\omega)E(\omega)E(\omega)E(\omega)\boldsymbol{x}$$

3rd Harmonic generation

$$\boldsymbol{P}^{(3)}(\omega) = 3\epsilon_0 \chi^{(3)}(\omega,-\omega,\omega)E(\omega)E(-\omega)E(\omega)\boldsymbol{x}$$

Optical Kerr Effect

$$\chi^{(3)}(\omega_1,\omega_2,\omega_3) = \frac{-N\gamma e^4}{\epsilon_0 m^3 \mathcal{D}(\omega_1+\omega_2+\omega_3)\mathcal{D}(\omega_1)\mathcal{D}(\omega_2)\mathcal{D}(\omega_3)}$$

3rd Order Nonlinear Polarization

Conclusion & Comments

- The macroscopic polarization induced inside the material is then given by the sum :

$$\begin{aligned}\boldsymbol{P}(z,t) &= \boldsymbol{P}^{(1)}(z,t) + \boldsymbol{P}^{(3)}(z,t)\\ &= \boldsymbol{P}(0) + \boldsymbol{P}(\omega)e^{-\imath(\omega t - kz)} + \boldsymbol{P}(3\omega)e^{-3\imath(\omega t - kz)} + CC.\end{aligned}$$

With : $\boldsymbol{P}^{(3)}(3\omega) = \epsilon_0 \chi^{(3)}(\omega,\omega,\omega)E(\omega)E(\omega)E(\omega)\boldsymbol{x}$ (Complex amplitudes)

$$\boldsymbol{P}^{(3)}(\omega) = 3\chi^{(3)}(\omega,-\omega,\omega)E(\omega)E(-\omega)E(\omega)\boldsymbol{x}$$

$$\chi^{(3)}(\omega_1,\omega_2,\omega_3) = \frac{-N\gamma e^4}{\epsilon_0 m^3 \mathcal{D}(\omega_1+\omega_2+\omega_3)\mathcal{D}(\omega_1)\mathcal{D}(\omega_2)\mathcal{D}(\omega_3)} \qquad \mathcal{D}(\omega) = \omega_0^2 - \omega^2 - \imath\alpha\omega$$

- For **non-resonant interactions** ($\omega \ll \omega_0$)
  $\Rightarrow$ **$\chi^{(3)}$ is real and independent of the frequency**

- Strong enhancement of the nonlinear susceptibility is expected once ω or 3ω (or both) is close to a material transition (@ω_0)

- Phase mismatching between the polarization component @ 3ω and the free propagative wave @ 3ω : **3k(ω) ≠ k(3ω)**