Ch. 1 INTRODUCTION TO NONLINEAR OPTICS

- Nonlinear regime - Order of magnitude
- Origin of the nonlinearities
  - Induced Dipole and Polarization
  - Description of the classical anharmonic oscillator model
  - Linear polarization and susceptibility
  - Nonlinear polarization and susceptibility
- Nonlinear interactions : a brief description
Induced dipole and macroscopic polarization

✓ **Microscopic scale**

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

- Simplistic model for an atom =
  - Nucleus (charge +)
  - Electronic cloud (charge -)

- 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 field = temporal deformation of the electronic cloud

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

\( E \) time

\( \vec{p} \) time

Induced Dipole

- time variation of the position of the center of mass : \( \Delta x(t) \)
- the electronic cloud is linked with the nucleus, presence of a restoring force (linear function of the displacement of the electrons)
- vibrating dipole (induced dipole):

*In a linear regime*: linear relation between the displacement of the electrons and the restoring force

**Induced dipole frequency** \( \omega = \) applied EM field frequency \( \omega \)

Abrev. : EM field means Electromagnetic Field
Induced dipole and macroscopic polarization

- **Macroscopic scale**

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

\[ \mathbf{E}(t) = A \cos(\omega t + kz) \]

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

Case of a z propagative EM wave with freq. \( \omega \)

Source term @ \( \omega \)

The polarization vector \( \mathbf{P}(t) \) acts as a source term in the wave equation

\[ \mathbf{P}(t) = N \times \mathbf{p} = \varepsilon_0 \chi^{(1)} \mathbf{E}(t) \]

Def.: LINEAR susceptibility of the medium

Polarisation = « macroscopic » source term
Classical anharmonic oscillator model

**Classical anharmonic oscillator - Description**

Induced dipole (microscopic quantity): \[ p(z, t) = -e \, x(z, t) \, \cdot \, \mathbf{E} \]

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

**Equation of motion**

\[ \frac{d^2 x}{dt^2} + \alpha \frac{dx}{dt} + \omega_0^2 x + \beta x^2 + \gamma x^3 + \cdots = \frac{-e}{m} \, x \cdot \mathbf{E}(z, t) \]
Classical anharmonic oscillator model

- Two applied EM wave with frequencies $\omega_1$ and $\omega_2$:

**LINEAR CASE** : « LINEAR » response of the vibrating dipole

Induced Dipole

Vibrating dipoles with frequencies = $\omega_1$ and $\omega_2$

A material with a collection of N dipoles in a linear vibrating regime = LINEAR MEDIUM
Classical anharmonic oscillator model

- Two applied EM wave with frequencies $\omega_1$ and $\omega_2$:

**NONLINEAR CASE**: « NONLINEAR » dependence of the restoring force with the displacement of the center of mass

\[ \text{Induced Dipole} \]

Vibrating dipoles with frequencies = $\omega_1$ and $\omega_2$ + harmonics

A material with a collection of N dipoles in a nonlinear vibrating regime = NONLINEAR MEDIUM
Classical anharmonic oscillator model

- **Equation of motion**

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

Damping term  | Restoring force  | Driven Coulomb force

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

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

\[x^{(1)} \gg x^{(2)} \gg x^{(3)}\]
**Linear polarization**

**Harmonic oscillator : equation of motion**

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

**Driven solution**

\[
x^{(1)}(z, t) = a(\omega)e^{-i(\omega t - k z)} + a(-\omega)e^{i(\omega t - k z)}
\]

**Induced dipole**

\[
p^{(1)}(z, t) = \alpha^{(1)}(\omega)A(\omega)e^{-i(\omega t - k z)} x + CC.
\]

**Linear polarizability**

(microscopic quantity)

\[
\alpha^{(1)} = \frac{e^2}{mD(\omega)} \quad D(\omega) = \omega_0^2 - \omega^2 - i\alpha\omega
\]

**Macroscopic Polarization**

**Linear susceptibility**

\[
\mathcal{P}^{(1)}(z, t) = \varepsilon_0\chi^{(1)}(\omega)E(\omega)e^{-i\omega t} + CC.
\]
Linear polarization

Harmonic oscillator: equation of motion

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

\[
P^{(1)}(z, t) = \epsilon_0 \chi^{(1)}(\omega) E(\omega) e^{-\omega t} + CC.
\]

Linear Susceptibility

\[
\chi^{(1)}(\omega) = \frac{Ne^2}{\epsilon_0 m (\omega_0^2 - \omega^2 - i\alpha \omega)}
\]

\[
\chi^{(1)}(\omega) = \chi'(\omega) + i\chi''(\omega)
\]

Dispersion

Absorption (or amplification)

Lorentzian line shape
2nd Order Nonlinear Polarization

Anharmonic oscillator: equation of motion

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

Driven solution

\[ x(z, t) = \lambda x^{(1)}(z, t) + \lambda^2 x^{(2)}(z, t) + \cdots \]

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

\[ x^{(2)}(z, t) = b(0) + b(2\omega)e^{-2i(\omega t - k z)} + b(-2\omega)e^{2i(\omega t - k z)} \]

\[
\begin{cases}
   b(0) = \frac{-2\beta e^2 |A|^2(\omega)}{m^2 \mathcal{D}(0)\mathcal{D}(\omega)\mathcal{D}(-\omega)} \\
   b(\pm 2\omega) = \frac{-\beta e^2 A^2(\pm \omega)}{m^2 \mathcal{D}(\pm 2\omega)\mathcal{D}(\pm \omega)\mathcal{D}(\pm \omega)}
\end{cases}
\]
2nd Order Nonlinear Polarization

Anharmonic oscillator: equation of motion

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

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

\[ P^{(2)}(2\omega) = \epsilon_0\chi^{(2)}(\omega, \omega)E(\omega)E(\omega)x \]

2nd order Nonlinear Susceptibility

\[ \chi^{(2)}(\omega_1, \omega_2) = \frac{N\alpha^{(2)}(\omega_1, \omega_2)}{\epsilon_0} \]

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

Optical RECTIFICATION
(induces static electric field)

2nd Harmonic Generation
creation of an electric field with
a 2\omega frequency component
Anharmonic oscillator: equation of motion

Conclusion & Comments

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

\[ P(z, t) = P^{(1)}(z, t) + P^{(2)}(z, t) \]

\[ = P^{(2)}(0) + P^{(1)}(\omega) e^{-i(\omega t - k z)} + P^{(2)}(2\omega) e^{-2i(\omega t - k z)} + C C' \]

With:

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

\[ P^{(2)}(2\omega) = \epsilon_0 \chi^{(2)}(\omega, \omega) E(\omega) E(\omega) x \]  

(Complex amplitudes)

• Phase mismatching between the polarization component @ 2\omega and the free propagative wave @ 2\omega:

wavevector related to P(2\omega) ≠ wavevector of E(2\omega)

\[ 2k(\omega) ≠ k(2\omega) \]

• Strong enhancement of the nonlinear susceptibility is expected once \( \omega \) or 2\( \omega \) (or both) is close to a material transition (@ \( \omega_0 \))

\[ \chi^{(2)}(\omega_1, \omega_2) = \frac{N e^3}{m^2 D(\omega_1 + \omega_2) D(\omega_1) D(\omega_2)} \]

\[ D(\omega) = \omega_0^2 - \omega^2 - i\omega \]
3rd Order Nonlinear Polarization

Anharmonic oscillator: equation of motion

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

Driven solution

\[
x(z, t) = \lambda x^{(1)}(z, t) + \lambda^2 x^{(2)}(z, t) + \lambda^3 x^{(3)}(z, t)
\]

Linear and Nonlinear Polarization

\[
\mathcal{P}(z, t) = \mathcal{P}^{(1)}(z, t) + \mathcal{P}^{(3)}(z, t)
\]

Case of a centro-symmetric material

\[
x^{(2)} = 0
\]

3rd Harmonic generation

\[
P^{(3)}(3\omega) = \epsilon_0 \chi^{(3)}(\omega, \omega, \omega) E(\omega) E(\omega) E(\omega) x
\]

Optical Kerr Effect

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

\[
\chi^{(3)}(\omega_1, \omega_2, \omega_3) = \frac{-N\gamma e^4}{\epsilon_0 m^3 D(\omega_1 + \omega_2 + \omega_3) D(\omega_1) D(\omega_2) D(\omega_3)}
\]
Conclusion & Comments

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

\[ P(z, t) = P^{(1)}(z, t) + P^{(3)}(z, t) \]
\[ = P(0) + P(\omega)e^{-i(\omega t - kz)} + P(3\omega)e^{-3i(\omega t - k_3z) + CC}. \]

With:

\[ P^{(3)}(3\omega) = \epsilon_0\chi^{(3)}(\omega, \omega, \omega)E(\omega)E(\omega)E(\omega)x \]

(Complex amplitudes)

\[ P^{(3)}(\omega) = 3\chi^{(3)}(\omega, -\omega, \omega)E(\omega)E(-\omega)E(\omega)x \]

• Phase mismatching between the polarization component @ 3\omega and the free propagative wave @ 3\omega:

\[ 3k(\omega) \neq k(3\omega) \]

• Strong enhancement of the nonlinear susceptibility is expected once \( \omega \) or 3\( \omega \) (or both) is close to a material transition (@\( \omega_0 \))

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

\[ D(\omega) = \omega_0^2 - \omega^2 - i\alpha\omega \]
Nonlinear interactions

✓ A short review:

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

\[ E(t) = A \cos(\omega t + k z) \]

\[ P(t) \propto \chi^{(1)} A \cos(\omega t + k z) \]

The polarization vector \( P(t) \) acts as a source term in the wave equation.

\[ \vec{P}(t) = N \times \vec{p} = \varepsilon_0 \chi^{(1)} \vec{E}(t) \]

Def.: LINEAR susceptibility of the medium
Nonlinear interactions

✓ **A short review:**

- **Case of a **NONLINEAR MEDIUM**: collection of \( N \) identical atoms

The nonlinear response of the medium can be expressed as

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

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

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

**Linear response**

**2nd Order** + **3rd Order**

**Nonlinear Response**

Considering a z propagative EM wave @ \( \omega \)

\[
E(t) = A \cos(\omega t + kz)
\]

**1st Order**

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

Source term @ \( \omega \)

**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 @ \( 2\omega \) !!!
A short review:

Case of a NONLINEAR MEDIUM: collection of N identical atoms

The nonlinear response of the medium can be expressed as:

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

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

Considering a z-propagative EM wave @ \( \omega \):

1st Order:

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

Source term @ \( \omega \)

2nd Order + 3rd Order Nonlinear Response

3rd Order:

\[ E(t) = A \cos(\omega t + kz) \]

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

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

Source term @ \( 3\omega \)

Third harmonic generation

Source term @ \( \omega \)

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**

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

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

\[ P_L(t) \propto \chi^{(1)} A \cos(\omega t + kz) \quad \text{LINEAR Regime} \]

Directly related to the refractive index

Considering a propagative EM wave @ \( \omega \)

\[ E(t) = A \cos(\omega t + kz) \]

\[ I \propto A^2 \quad \text{Wave intensity} \]

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

⇒ 2nd order nonlinearities
• optical rectification: induces a static electric field
• 2nd harmonic generation: generate a EM wave @ $2\omega$
• Sum and Difference frequency generation: generate a EM wave @ $\omega_1 \pm \omega_2$, fluorescence, amplification and parametric oscillation

⇒ 3rd order nonlinearities
• Optical Kerr effect: nonlinear refractive index change
• Solitons
• Self-phase modulation
• Four-wave mixing: nonlinear interaction of 4 degenerates or non-degenerates waves
• Raman scattering
• Brillouin scattering
Example: Four-wave mixing

Nonlinear interaction of 3 waves with 3 different frequencies

\[ E_1(t) = A_1 \cos(\omega_1 t + k_1 z) \]
\[ E_2(t) = A_2 \cos(\omega_2 t + k_2 z) \]
\[ E_3(t) = A_3 \cos(\omega_3 t + k_3 z) \]

\[ P_{NL}(t) \propto \chi^{(3)} E_1(t) E_2(t) E_3(t) \]

Frequency components of the nonlinear polarization:

- \( \omega_1 + \omega_2 + \omega_3 \)
- \( \omega_1 + \omega_2 - \omega_3 \)
- \( \omega_1 - \omega_2 + \omega_3 \)
- \( \omega_1 - \omega_2 - \omega_3 \)
- \( \omega_3 \)
- \( \omega_2 \)
- \( \omega_1 \)
Example : Four-wave mixing

Example : Source term @ $\omega_4 = \omega_1 + \omega_2 - \omega_3$

Assumption : low dispersive medium / The refractive indices @ $\omega_1, \omega_2, \omega_3, \omega_4$ are equals $\Rightarrow$ the polarization @ $\omega_4$ radiates in phase with a z propagative field @ $\omega_4 = \langle$ phase matching condition $\rangle$

$k_4 = k_1 = k_2 = k_3$

$P_{NL}(t) \propto \chi^{(3)} A^3 \cos\left( (\omega_1 + \omega_2 - \omega_3) t + (k_1 + k_2 - k_3) z \right)$

$E_4(t) = A_4 \cos(\omega_4 t + k_4 z)$

Once the phase matching condition is fulfilled, the in-phase array of dipoles contributes to efficiently generate a propagative field @ $\omega_4$
**Example : Four-wave mixing**

**Example :** Source term @ \( \omega_4 = \omega_1 + \omega_2 - \omega_3 \)

**Assumption :** dispersive medium / The refractive indices @ \( \omega_1, \omega_2, \omega_3, \omega_4 \) are no more equals \( \Rightarrow \) the radiating polarization @ \( \omega_4 \) is no more in phase with the z propagative field @ \( \omega_4 = \text{« NO phase matching condition »} \)

\[
P_{NL}(t) \propto \chi^{(3)} A^3 \cos \left( (\omega_1 + \omega_2 - \omega_3) t + (k_1 + k_2 - k_3) z \right)
\]

\[
E_4(t) = A_4 \cos (\omega_4 t + k_4 z)
\]

The EM field @ \( \omega_4 \) can not be efficiently generated because of a strong phase mismatch between the source term and the propagative wave @ \( \omega_4 \)

N. Dubreuil - **Nonlinear Optics**