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2328-2

Preparatory School to the Winter College on Optics and the Winter College on Optics: Advances in Nano-Optics and Plasmonics

6 - 17 February, 2012

Optical nonlinear processes

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Nonlinear Optics

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OUTLINE

Linear and Nonlinear Systems

Linear susceptibility: dispersion

Nonlinear Susceptibility

Nonlinear Optical Interactions

Second harmonic

Other nonlinear processes

Linear and Nonlinear Systems

A **linear system** is defined as one which has a response proportional to external influence and has a well-known property, i.e. if influences, F1, F2 …., Fn are applied simultaneously, the response produced is the sum of the responses that would be produced if the influences were applied separately.

A **nonlinear system** is one in which the response is not strictly proportional to the influence and the transfer of energy from one influence to another can occur.

If the **influences** are periodic in time, the response of a nonlinear system can contain frequencies different from those present in the influences. However, the point to emphasize here is that, as well as the generation of new frequencies, nonlinear optics provides the ability to control light with light and so to transfer information directly from one beam to another without the need to resort to electronics.

Traditionally, nonlinear optics has received a phenomenological approach in terms of **the effect of an electric field on the polarization within a material**.

MAXWELL EQUATIONS

Electromagnetic processes are described by Maxwell's equations which constitute a set of linear equations. In SI units:

$$\nabla \cdot \mathbf{D} = \rho \qquad \operatorname{div} \mathbf{D} = \rho$$

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} \qquad \operatorname{rot} \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}$$

$$\nabla \cdot \mathbf{B} = 0 \qquad \operatorname{div} \mathbf{B} = 0$$

$$\nabla \times \mathbf{H} = \frac{\partial \mathbf{D}}{\partial t} + \mathbf{J} \qquad \operatorname{rot} \mathbf{H} = \frac{\partial \mathbf{D}}{\partial t} + \mathbf{J}$$

where **E** and **B** are the electric and magnetic fields. The displacement fields **D** and **H** arise from the external charge and conduction current densities ρ and **J**. In most cases of interest in nonlinear optics, $\rho = 0$ and $J = 0$.

'Constitutive relations' connect the charge and current distributions within the medium and the displacement fields to the electric and magnetic fields.

$$\mathbf{D} = \mathbf{P} + \varepsilon_0 \mathbf{E} = \varepsilon \mathbf{E} \qquad \mathbf{B} = \mu \mathbf{H}$$

where **P** is the induced polarization in the medium resulting from the field E, ε is defined as a dielectric constant and ε_0 is the permittivity of free space (8.85×10^{-12} F m^{-1} in MKS units). Optical materials are mostly **non magnetic** $\mu = \mu_r \mu_0 = \mu_0$.

LINEAR THEORY

Usually one assumes a linear response of a dielectric material to an external field

$$\mathbf{P} = \varepsilon_0 \chi \mathbf{E}$$

Where **P** is the vector representing the electric dipole moment per unit volume induced by the external electric field **E**, ε_0 is vacuum permittivity and χ is a quantity characteristics of the considered material with no dimensions, called **electric susceptibility**.

In general χ is a tensor

$$\begin{vmatrix} P_x \\ P_y \\ P_z \end{vmatrix} = \varepsilon_0 \begin{vmatrix} \chi_{xx} & \chi_{xy} & \chi_{xz} \\ \chi_{yx} & \chi_{yy} & \chi_{yz} \\ \chi_{zx} & \chi_{zy} & \chi_{zz} \end{vmatrix} \begin{vmatrix} E_x \\ E_y \\ E_z \end{vmatrix}$$

The symmetry properties of the material indicate which ones of the χ_{ij} coefficients are zero.

Alternatively

$$\begin{vmatrix} D_x \\ D_y \\ D_z \end{vmatrix} = \begin{vmatrix} \varepsilon_{xx} & \varepsilon_{xy} & \varepsilon_{xz} \\ \varepsilon_{yx} & \varepsilon_{yy} & \varepsilon_{yz} \\ \varepsilon_{zx} & \varepsilon_{zy} & \varepsilon_{zz} \end{vmatrix} \begin{vmatrix} E_x \\ E_y \\ E_z \end{vmatrix}$$

HOMOGENEOUS MATERIALS

ε not depending on space

$$\operatorname{div}\varepsilon\mathbf{E}=\rho \qquad\qquad \operatorname{div}\mathbf{E}=\frac{\rho}{\varepsilon}$$

$$\operatorname{rot}\mathbf{E}=-\frac{\partial \mathrm{B}}{\partial t} \qquad \Rightarrow \qquad \operatorname{rot}\mathbf{E}=-\frac{\partial \mathbf{B}}{\partial t}$$

$$\operatorname{div}\mathbf{B}=0 \qquad\qquad \operatorname{div}\mathbf{B}=0$$

$$\operatorname{rot}\left(\frac{\mathbf{B}}{\mu}\right)=\left(\mathbf{j}+\frac{\partial \varepsilon\mathbf{E}}{\varepsilon t}\right) \qquad \operatorname{rot}\mathbf{B}=\mu\left(\mathbf{j}+\frac{\partial \varepsilon\mathbf{E}}{\partial t}\right)$$

If $\rho=0$ $\mathbf{j}=0$

$$\operatorname{div}\mathbf{E}=0 \qquad\qquad \operatorname{div}\mathbf{B}=0$$

$$\operatorname{rot}\mathbf{E}=-\frac{\partial \mathbf{B}}{\partial t} \qquad\qquad \operatorname{rot}\mathbf{B}=\mu\varepsilon\frac{\partial \mathbf{E}}{\partial t}$$

WAVE EQUATION

Starting from Maxwell's Eqs

$$\begin{cases} \operatorname{div}\vec{E} = 0 \\ \operatorname{rot}\vec{E} = -\dfrac{\partial \vec{B}}{\partial t} \\ \operatorname{div}\vec{B} = 0 \\ \operatorname{rot}\vec{B} = \mu_0 \dfrac{\partial \vec{D}}{\partial t} \end{cases}$$

$$\operatorname{rot}\operatorname{rot}\mathbf{E} = -\nabla^2\mathbf{E} + \operatorname{grad}\operatorname{div}\mathbf{E}$$

$$= -\frac{\partial}{\partial t}\operatorname{rot}\mathbf{B}$$

$$-\nabla^2\mathbf{E} + \operatorname{grad}\operatorname{div}\mathbf{E} = -\mu_0 \frac{\partial}{\partial t}\frac{\partial \mathbf{D}}{\partial t} = -\mu_0 \frac{\partial^2 \mathbf{D}}{\partial t^2}$$

$$\nabla^2\mathbf{E} = \left(\frac{\partial^2 \mathbf{E}_x}{\partial x^2} + \frac{\partial^2 \mathbf{E}_x}{\partial y^2} + \frac{\partial^2 \mathbf{E}_x}{\partial z^2}\right)\mathbf{i} + \left(\frac{\partial^2 \mathbf{E}_y}{\partial x^2} + \frac{\partial^2 \mathbf{E}_y}{\partial y^2} + \frac{\partial^2 \mathbf{E}_y}{\partial z^2}\right)\mathbf{j} + \left(\frac{\partial^2 \mathbf{E}_z}{\partial x^2} + \frac{\partial^2 \mathbf{E}_z}{\partial y^2} + \frac{\partial^2 \mathbf{E}_z}{\partial z^2}\right)\mathbf{k}$$

$$\nabla^2\mathbf{E} = -\mu_0\varepsilon\frac{\partial^2 \mathbf{E}}{\partial t^2} = \frac{1}{v^2}\frac{\partial^2 \mathbf{E}}{\partial t^2}$$

$$v = \frac{c}{\sqrt{\varepsilon_r}} = \frac{c}{n} \qquad c = \frac{1}{\sqrt{\varepsilon_0\mu_0}}$$

refractive index

$$n = \sqrt{\varepsilon_r}$$

DISPERSION

Experimentally the refractive index is a function of wavelength (frequency)

$$n(\lambda) = \sqrt{\varepsilon_r(\lambda)} \qquad \varepsilon_r(\lambda) = 1 + \chi(\lambda)$$

This phenomenon is called DISPERSION.

The polarization in a material medium can be explained considering the electrons tied to the atoms as harmonic oscillators.

Nucleus: ~2000 electron mass, i.e., infinite mass

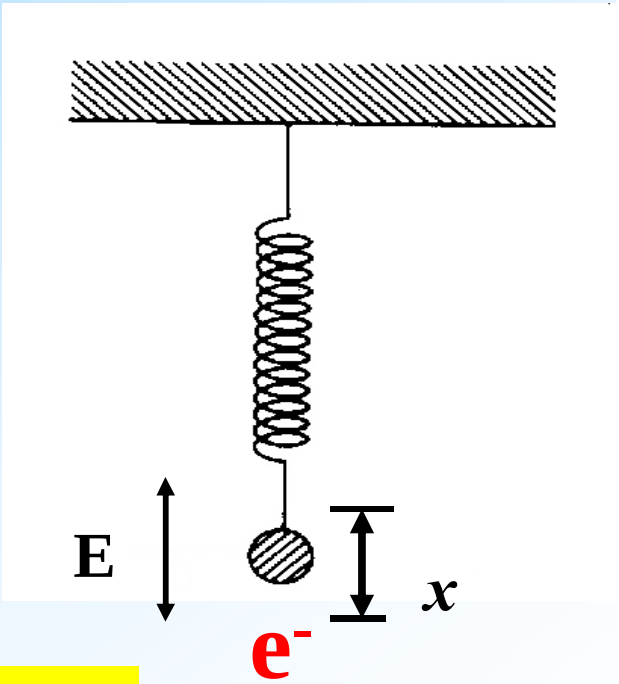
DISPERSION

$$m\ddot{x} + \beta\dot{x} + kx = -eE_0e^{i\omega t}$$ (one-dimensional model)

Damping **Restoring Force** **Driving Force**

From the solution:

$$x = \frac{-eE_0e^{i\omega t}}{m(\omega_0^2 - \omega^2 + i\omega\gamma)} \qquad \gamma = \frac{\beta}{m} \qquad \omega_0^2 = \frac{k}{m}$$



the induced moment is calculated:

$$p = -ex = \frac{e^2}{m\left(\omega_0^2 - \omega^2 + i\omega\gamma\right)} \cdot E_0e^{i\omega t}$$

For N oscillators per volume unit, the polarization is:

$$P = N \cdot p = \frac{N \cdot e^2}{m\left(\omega_0^2 - \omega^2 + i\omega\gamma\right)} E_0 e^{i\omega t}$$

Calling $\alpha = \dfrac{\mathrm{e}^2}{\mathrm{m}(\omega_0^2 - \omega^2 + \mathrm{i}\gamma\omega)} \equiv$ atomic polarizability

➔ $\mathrm{P} = \mathrm{N}\alpha\mathrm{E} = \varepsilon_0\chi\mathrm{E}$ $\qquad \mathrm{E} = \mathrm{E}_0\mathrm{e}^{\mathrm{i}\omega t}$ $\qquad \chi = \dfrac{\mathrm{N}\alpha}{\varepsilon_0}$

where χ is the electric susceptibility.

$$\varepsilon_0\varepsilon_\mathrm{r} = \varepsilon_0(1+\chi) = \varepsilon_0\left(1 + \frac{\mathrm{N}\alpha}{\varepsilon_0}\right)$$

$$\mathrm{n}^2 = 1 + \chi = 1 + \frac{\mathrm{N}\alpha}{\varepsilon_0} \qquad \mathrm{n} = \sqrt{\varepsilon_\mathrm{r}}$$

$$n^2 = 1 + \frac{Ne^2}{\varepsilon_0 m(\omega_0^2 - \omega^2 + i\gamma\omega)}$$

If the second term is lower than 1 (as it happens in gases):

$$n \cong 1 + \frac{Ne^2}{2\varepsilon_0 m(\omega_0^2 - \omega^2 + i\gamma\omega)}$$

In the expression n comes out to be a complex number.

ABSORPTION

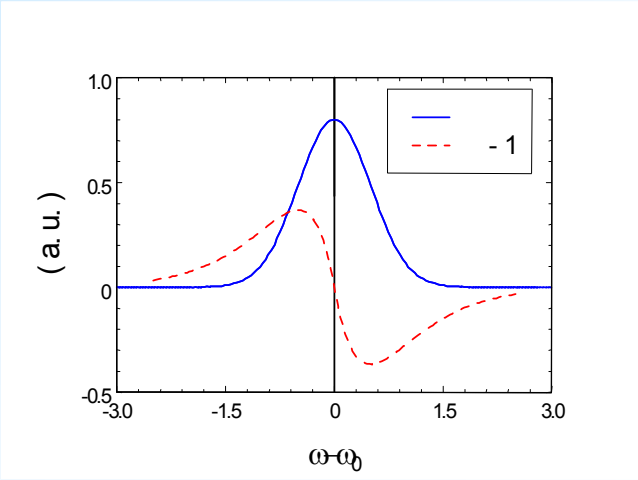
The term $i\gamma\omega$ is responsible for absorption. The complex index can be written as

$$n = \tilde{n} - i\tilde{k} = 1 + \frac{Ne^2(\omega_0^2 - \omega^2)}{2\varepsilon_0 m\left[\left(\omega_0^2 - \omega^2\right)^2 + \gamma^2\omega^2\right]} - i\frac{Ne^2\gamma\omega}{2\varepsilon_0 m\left[\left(\omega_0^2 - \omega^2\right)^2 + \gamma^2\omega^2\right]}$$

If we consider a plane wave

where $E = A\exp\left[i(\omega t - kz)\right]$

$$k = \omega\sqrt{\mu\varepsilon} = \frac{\omega}{c}n = \frac{2\pi}{\lambda}n$$



we see that, substituting the complex refractive index, one has

$$k = \frac{2\pi}{\lambda}(\tilde{n} - i\tilde{k})$$

which gives

$$E = A\exp\left[i\left(\omega t - \frac{2\pi\tilde{n}}{\lambda}z\right)\right]\exp\left(-\frac{2\pi\tilde{k}}{\lambda}z\right)$$

The last exponential represents a term of attenuation. The attenuation coefficient may be defined from:

$$-\alpha = \frac{1}{I}\frac{dI}{dz} \qquad I(z) = |E|^2 = I_{(0)}e^{-\alpha z}$$

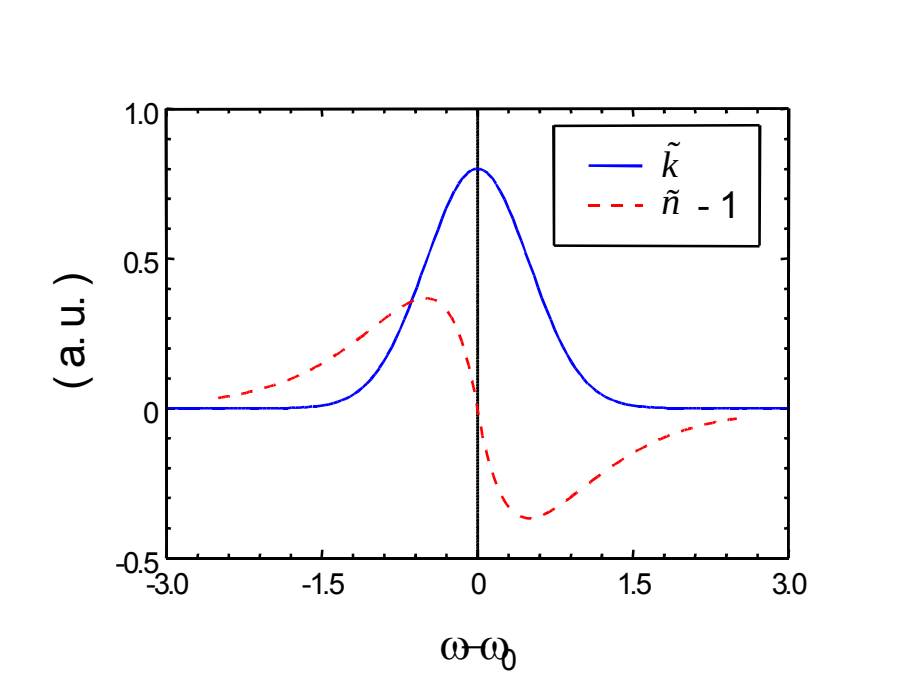
By comparison with the previous equation

$$E = A\exp\left[i\left(\omega t - \frac{2\pi\tilde{n}}{\lambda}z\right)\right]\exp\left(-\frac{2\pi\tilde{k}}{\lambda}z\right)$$

$$\alpha = \frac{4\pi}{\lambda}\tilde{k}$$

It can be noticed that a small value of $\tilde{k}$ leads to an elevated attenuation.

$\tilde{k} = 0.0001$ and $\lambda = 0.5\mu m$ gives $\alpha = 25\ \mathrm{cm}^{-1}$.



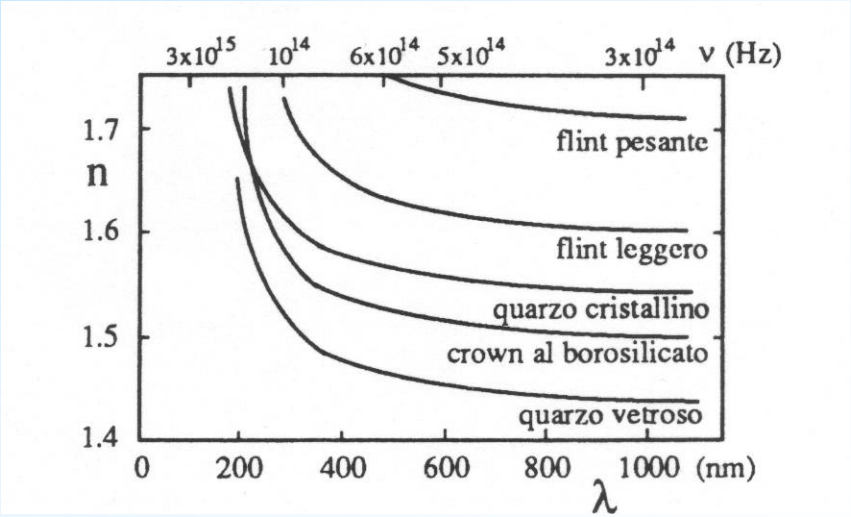
Both $\tilde{n}$ and $\tilde{k}$ are functions of the frequency.

Substance	n (for yellow light)	$\sqrt{\varepsilon_r}$ (static value)
Air (1 atm)	1.0002926	1.0002925
CO (1 atm)	1.00045	1.0005
Polistyren	1.59	1.6
Glass	1.5 ÷ 1.7	2 ÷ 3
Fused quartz	1.46	1.94
Water	1.33	9
Ethanol	1.36	5

Table I

Values of n and ε_r for some materials

n as a function of λ for some materials

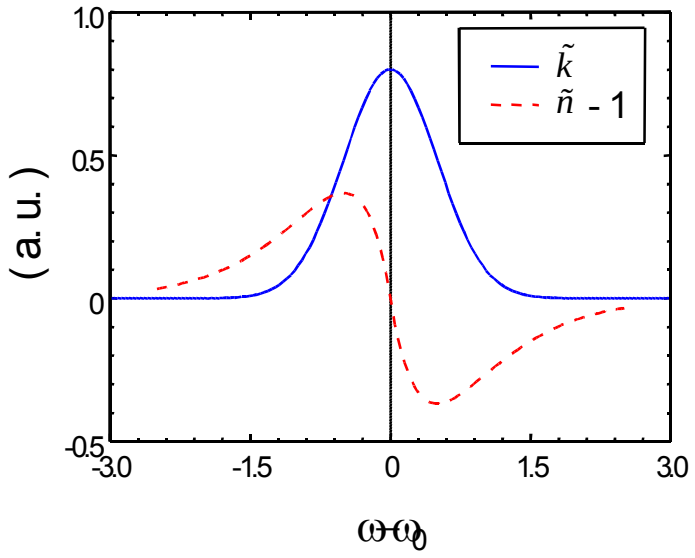


$\tilde{n} > 1$ for $\omega < \omega_0$ **normal dispersion**

At resonance ($\omega = \omega_0$)

the slope of $\tilde{n}$ is negative **anomalous dispersion**

Absolute refraction index at 20°C for the line D of Sodium (λ=5890 Å)					
Solids	n	Liquids	n	Gas	n
Canadian balsam	1.528	Acetone	1.359	Carbon dioxide	1.000448
Calcite	1.658	Water	1.333	Air	1.000292
Dispersive Crown	1.520	Ethanol	1.361	Nitrogen	1.000296
Heavy Flint	1.650	Benzene	1.502	Helium	1.000036
Amorphous quartz	1.458	Etere etilico	2.352	Hydrogen	1.000132
Heavy glass	1.970	Solfuro di Carbonio	1.627	Oxygen	1.000271



METALS

In a metal the electrons are free and they do not oscillate around the atoms. **Therefore k = 0 and $\omega_0 = 0$.**

In the equation for n^2 it is sufficient to put $\omega_0 = 0$.

$$n^2 = 1 - \frac{Ne^2}{\varepsilon_0 m(\omega^2 - i\gamma\omega)}$$

$N \equiv$ density of electrons

If $\gamma << \omega$

$$n^2 \approx 1 - \frac{\omega_p^{\,2}}{\omega^2}$$

$$\omega_p^{\,2} = \frac{Ne^2}{\varepsilon_0 m}$$

Frequency of plasma

For Al, Cu, Au, Ag N ~ 10^{23} cm^{-3} and $\omega_P \sim 2.10^{16}$ s^{-1}.

For $\omega > \omega_P$ n is real and the waves propagate freely.

For $\omega < \omega_P$ n is pure imaginary and the field is exponentially attenuated with the distance from the surface. Therefore the radiation is reflected from the surface.

Therefore, for visible radiation and infrared $\omega < \omega_P$ and n is imaginary. In general, n is complex because there is γ:

$$1-n^2 = \frac{Ne^2}{\omega\varepsilon_0 m(\omega - i\gamma)}\frac{(\omega + i\gamma)}{(\omega + i\gamma)} = \frac{Ne^2}{\omega\varepsilon_0 m\left(\omega^2+\gamma^2\right)} + i\gamma\frac{Ne^2}{\omega\varepsilon_0 m\left(\omega^2+\gamma^2\right)} =$$

$$\frac{\omega_p}{\left(\omega^2+\gamma^2\right)} + i\frac{\omega_p}{\left(\omega^2+\gamma^2\right)}\frac{\gamma}{\omega}$$

NONLINEAR SUSCEPTIBILITY

Dipole moment per unit volume or polarization in the linear case

$$P_i = P_i^0 + \chi_{ij} E_j$$

The general form of polarization in a nonlinear medium is

$$P_i = P_i^0 + \chi_{ij}^{(1)} E_j + \chi_{ijk}^{(2)} E_j E_k + \chi_{ijkl}^{(3)} E_j E_k E_l + \cdots$$

NONLINEAR SUSCEPTIBILITY

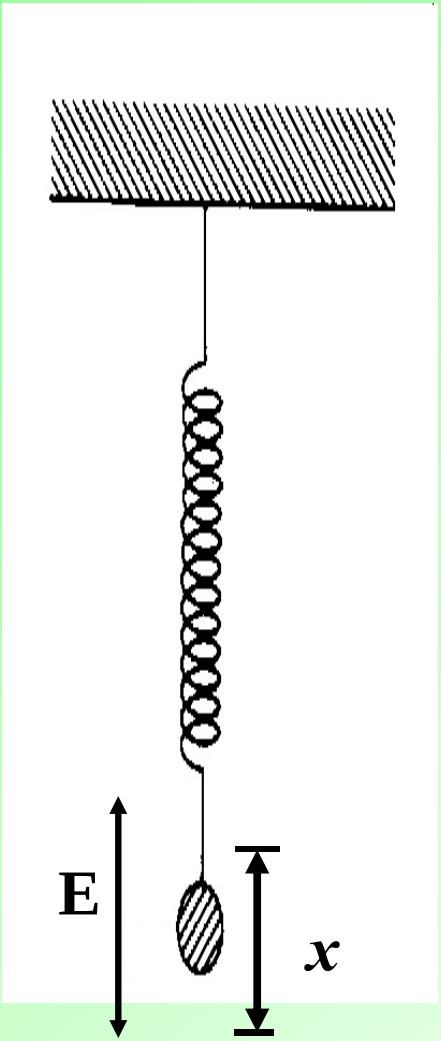
In some cases also the magnetic field is important and quadrupole terms

In these cases the general form of polarization in a nonlinear medium is

$$P_i = P_i^0 + \chi_{ij}^{(1)} E_j + \chi_{ijk}^{(2)} E_j E_k + \chi_{ijkl}^{(3)} E_j E_k E_l + \cdots$$

$+ \chi_{ijk}^{(2)} E_j B_k + \chi_{ijkl}^{(2)} (\nabla \cdot E) E + ..$

JUSTIFICATION OF THE PRESENCE OF A NONLINEAR RESPONSE



If the force exercised by the electric field of the wave becomes comparable with the Coulomb's force between the electron and the nucleus, the oscillator is perturbed (**anharmonic oscillator**) and, at the lower level of the perturbation, we can write:

$$\ddot{x}(t) + \sigma\dot{x}(t) + \omega_0^2 x(t) + Dx^2(t) = -(e/m)E(t) \qquad (4)$$

The solution of eq.(4) can express as the sum of two terms

$$x(t) = x^{(1)}(t) + x^{(2)}(t) \qquad (5)$$

in which $x^{(1)}(t)$ is obtained solving eq.(4) without the anharmonic term, whereas $x^{(2)}(t)$ is considered a small correction of the solution at the first order $x^{(1)}(t)$ and is obtained utilizing $x^{(1)}(t)$ in the anharmonic term

$$\ddot{x}^{(2)}(t) + \sigma\dot{x}^{(2)}(t) + \omega_0^2 x^{(2)}(t) = -\frac{eE(t)}{m} - D\left[x^{(1)}(t)\right]^2. \qquad (6)$$

In this way, considering the case in which the forcing electric field is formed by the sum of two fields at different frequencies

$$E(t) = E_1 \cos\omega_1 t + E_2 \cos\omega_2 t = \frac{1}{2}\left[E_1 e^{-j\omega_1 t} + E_2 e^{-j\omega_2 t} + c.c.\right] \qquad (7)$$

We have the solution at the **first order**

$$x^{(1)}(t)=\frac{1}{2}\left[x^{(1)}(\omega_1)e^{-j\omega_1 t}+x^{(1)}(\omega_2)e^{-j\omega_2 t}+\text{c.c.}\right] \qquad (8)$$

and subsequently the solution at the **second order**, solving eq.(6) with the use of (8) is

$$x^{(2)}(t)=\frac{1}{2}[x^{(2)}(\omega_1+\omega_2)e^{-j(\omega_1+\omega_2)t}+x^{(2)}(\omega_1-\omega_2)e^{-j(\omega_1-\omega_2)t}+$$
$$+x^{(2)}(2\omega_1)e^{-j2\omega_1 t}+x^{(2)}(2\omega_2)e^{-j2\omega_2 t}+\text{c.c}] \qquad (9)$$

in which

$$x^{(2)}(\omega_1\pm\omega_2)=-\frac{1}{2}\frac{D(e/m)^2}{(\omega_0^2-\omega_1^2+j\sigma\omega_1)(\omega_0^2-\omega_2^2+j\sigma\omega_2)}\cdot\frac{E_1E_2}{\left[\omega_0^2-(\omega_1\pm\omega_2)^2+j\sigma(\omega_1\pm\omega_2)\right]}$$

$$x^{(2)}(2\omega_k)=-\frac{1}{2}\frac{D(e/m)^2\cdot E_k^2}{(\omega_0^2-\omega_k^2+j\sigma\omega_k)^2(\omega_0^2-4\omega_k^2+j\sigma\omega_k)};\quad k=1,2. \qquad (10)$$

Therefore the solution of the second order brings to the generation of **oscillations at a frequency different from the ones of the forcing field**. In particular, it is possible to have frequencies equal to the **sum** or to the **difference** of the field frequencies or to the double (second harmonic). Moreover, we emphasize that the previous formulas remain valid also if just a single forcing field ω is present. In this case $x^{(2)}(t)$ will be the sum of a second harmonic term (2ω) with a null pulsation term (term of optical rectification).

Now, remembering the expression for the polarization of the medium, we can write

$$P(t) = -Ne\left[x^{(1)}(t) + x^{(2)}(t)\right] \qquad (11)$$

where N is the number of dipoles for volume unit; that is

$$P(t) = P_L(t) + P_{NL}(t) \qquad (12)$$

Which, compared with (10)

$$x^{(2)}(\omega_1 \pm \omega_2) = -\frac{1}{2}\frac{D(e/m)^2}{\left(\omega_0^2 - \omega_1^2 + j\sigma\omega_1\right)\left(\omega_0^2 - \omega_2^2 + j\sigma\omega_2\right)} \cdot \frac{E_1 E_2}{\left[\omega_0^2 - (\omega_1 \pm \omega_2)^2 + j\sigma(\omega_1 \pm \omega_2)\right]}$$

$$x^{(2)}(2\omega_k) = -\frac{1}{2}\frac{D(e/m)^2 \cdot E_k^2}{\left(\omega_0^2 - \omega_k^2 + j\sigma\omega_k\right)^2\left(\omega_0^2 - 4\omega_k^2 + j\sigma\omega_k\right)}; \quad k = 1,2. \qquad (10)$$

permits to write

$$P_L = \varepsilon_0 \chi^{(1)} E$$

$$P_{NL} = \chi^{(2)} E \cdot E. \qquad (13)$$

SECOND HARMONIC PRODUCTION

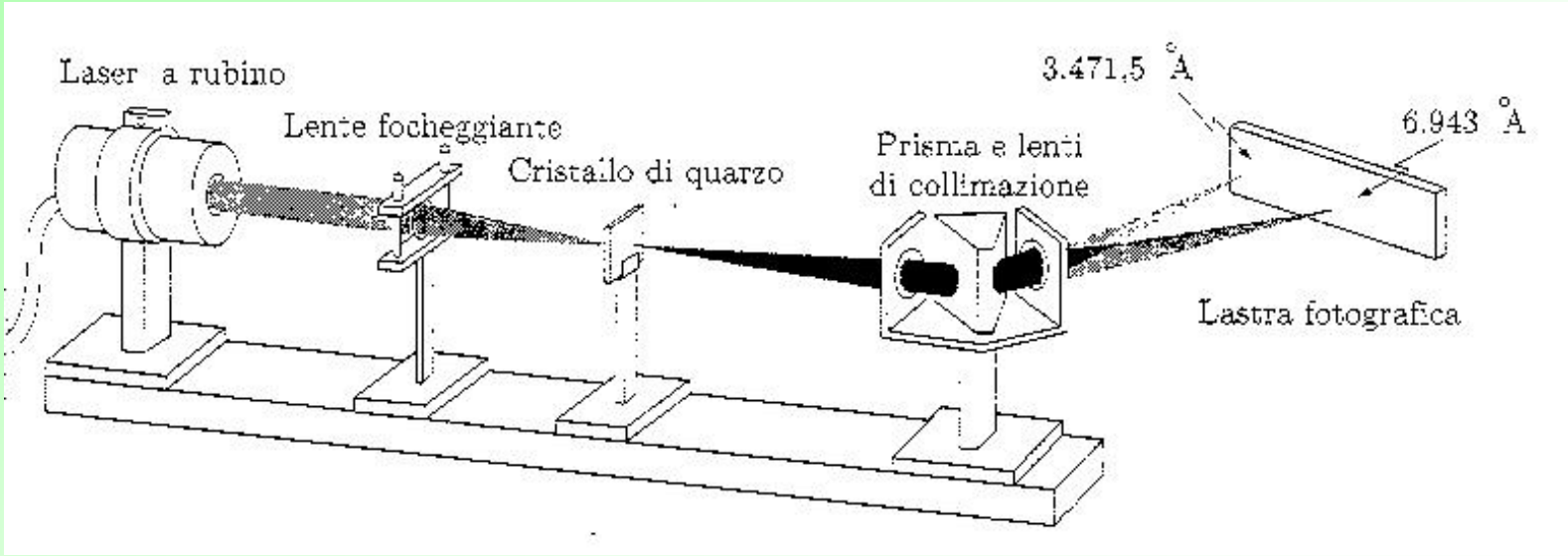
The nonlinear properties in the optical region have been demonstrated for the first time in **1961** by Franken et al. during an experiment of **second harmonic generation**. Sending red light of a ruby laser (λ = 6.943 Å) onto a crystal of quartz, they observed ultraviolet light.

VOLUME 7, NUMBER 4 PHYSICAL REVIEW LETTERS AUGUST 15, 1961

GENERATION OF OPTICAL HARMONICS*

P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich
The Harrison M. Randall Laboratory of Physics, The University of Michigan, Ann Arbor, Michigan

(Received July 21, 1961)



To describe the phenomenon, it is necessary to observe that in many crystal materials the nonlinear polarization depends on the **direction of propagation**, on the polarization of the electric field and on the orientation of the optical axis of the crystal. Since in such materials the vectors **P** and **E** are not necessarily parallel, the coefficient χ is a tensor. The second order polarization can be written as

$$P_i^{(2)} = \sum_{j,k} d_{ijk} E_j E_k \qquad (14)$$

where i, j, k represent the coordinates x, y, z. The main part of the coefficients d_{ijk}, however, are usually zero and so only a few of them must be considered.

Only the **non-centrosymmetric crystals** can have a non null tensor d_{ijk}. In facts, let us consider an isotropic crystal. In this case d_{ijk} is independent from the direction and therefore it is constant. If now we invert the direction of the electric field, also the polarization must change sign, that is

$$-P_i^{(2)} = \sum d_{ijk}(-E_j)(-E_k) = \sum d_{ijk} E_j E_k = +P_i^{(2)}.$$

It is clear that, not being able to be $-P_i^{(2)} = +P_i^{(2)}$, d_{ijk} must be null.

Moreover, in materials for which $d \neq 0$, since no physical meaning can be assigned to an exchange of E_j with E_k, it must be $d_{ijk} = d_{ikj}$.

Now if we consider the Maxwell equations writing

$$\mathrm{D} = \varepsilon_0 \mathrm{E} + \mathrm{P} \qquad (15)$$

we have

$$\mathrm{rot}\,\mathrm{B} = \mu \mathrm{j} + \mu \frac{\partial \mathrm{D}}{\partial \mathrm{t}} = \mu \mathrm{j} + \mu \varepsilon_0 \frac{\partial \mathrm{E}}{\partial \mathrm{t}} + \mu \frac{\partial \mathrm{P}}{\partial \mathrm{t}}$$

$$\mathrm{rot}\,\mathrm{E} = -\frac{\partial \mathrm{B}}{\partial \mathrm{t}}. \qquad (16)$$

The polarization can be written as the sum of a linear term plus a nonlinear one

$$\mathrm{P} = \varepsilon_0 \chi_\mathrm{L} \mathrm{E} + \mathrm{P}_\mathrm{NL} \qquad (17)$$

where, in case of materials with second order nonlinearity is, f.e.

$$\left(\mathrm{P}_\mathrm{NL}\right)_\mathrm{i} = \sum \mathrm{d}_{\mathrm{ijk}} \mathrm{E}_\mathrm{j} \mathrm{E}_\mathrm{k}. \qquad (18)$$

So eq.(16) can be written, assuming j = 0

$$\mathrm{rot}\,\mathrm{B} = \mu\frac{\partial \varepsilon \mathrm{E}}{\partial t} + \mu\frac{\partial \mathrm{P_{NL}}}{\partial t} \qquad (19)$$

from which

$$\nabla^2 \mathrm{E} = \mu\varepsilon\frac{\partial^2 \mathrm{E}}{\partial t^2} + \mu\frac{\partial^2 \mathrm{P_{NL}}}{\partial t^2}. \qquad (20)$$

If we consider the unidimensional case of propagation along a direction z, we have

$$\frac{\partial^2 \mathrm{E_i}}{\partial z^2} = \mu\varepsilon\frac{\partial^2 \mathrm{E_i}}{\partial t^2} + \mu\frac{\partial^2 (\mathrm{P_{NL}})_i}{\partial t^2}. \qquad (21)$$

Let us consider now three monochromatic fields with frequencies ω_1, ω_2, ω_3 using the complex notation

$$E_i^{(\omega_1)}(z,t) = \frac{1}{2}\left[E_{1i}(z) e^{j(\omega_1 t - k_{1z})} + c.c. \right]$$

$$E_k^{(\omega_2)}(z,t) = \frac{1}{2}\left[E_{2k}(z) e^{j(\omega_2 t - k_{2z})} + c.c. \right]$$

$$E_j^{(\omega_3)}(z,t) = \frac{1}{2}\left[E_{3j}(z) e^{j(\omega_3 t - k_{3z})} + c.c. \right] \qquad (22)$$

where the indices i, j, k represent the components x or y.

$$(P_{NL})_i = \sum d_{ijk} E_j E_k . \qquad (18)$$

The polarization at frequency $\omega_1 = \omega_3 - \omega_2$, for example, from (18) and from (22) results

$$P_i^{(\omega_1)} = \frac{1}{2} \sum_{j,k} d_{ijk} E_{3j}(z) E_{2k}^*(z) e^{j[(\omega_3 - \omega_2)t - (k_3 - k_2)z]} + c.c. \qquad (23)$$

Substituting eqs.(22) into (21) for the component E_{1i}, it is necessary to calculate

$$\frac{\partial^2 E^{(\omega_1)}}{\partial z^2} = \frac{1}{2}\frac{\partial^2}{\partial z^2}\left[E_{1i}(z)e\left(\omega_1 t - k_1 z\right) + c.c.\right]. \quad (24)$$

If we assume

$$\frac{dE_{1i}}{dz}k_1 \gg \frac{d^2E_{1i}}{dz^2} \quad (25)$$

we have

$$\frac{\partial^2 E_i^{(\omega_1)}}{\partial z^2} = -\frac{1}{2}\left[k_1^2 E_{1i}(z) + 2jk_1\frac{dE_{1i}(z)}{dz}\right]e^{j(\omega_1 t - k_1 z)} + c.c. \quad (26)$$

with similar expressions for

$$\frac{\partial^2 E_j^{(\omega_2)}}{\partial z^2} \quad \text{and} \quad \frac{\partial^2 E_k^{(\omega_3)}}{\partial z^2}.$$

Finally, substituting (26) and (23) into (21) we have

$$\frac{dE_{1i}(z)}{dz} = -j\frac{\omega_1}{2}\sqrt{\frac{\mu_0}{\varepsilon_1}}\sum d_{ijk}E_{3j}E_{2k}^{*}e^{-j(k_3-k_2-k_1)z} + c.c. \qquad (27)$$

and in analogous way

$$\frac{dE_{2k}^{*}}{dz} = \frac{j\omega_2}{2}\sqrt{\frac{\mu_0}{\varepsilon_2}}\sum d_{ijk}E_{1i}E_{3j}^{*}e^{-j(k_1-k_3+k_2)z} + c.c.$$

$$\frac{dE_{3j}}{dz} = -j\frac{\omega_3}{2}\sqrt{\frac{\mu_0}{\varepsilon_3}}\sum d_{ijk}E_{1i}E_{2k}e^{-j(k_1+k_2-k_3)z} + c.c.. \qquad (28)$$

The **second harmonic generation** is obtained immediately from (27) and (28) for the case of $\omega_1 = \omega_2$ and $\omega_3 = 2\omega_1$. Therefore it is enough to consider only, f.e., (27) and the last one of (28).

To further simplify the analysis we can assume that the power lost by the frequency ω_1 (fundamental) is negligible, and therefore

$$\frac{dE_{1i}}{dz} \simeq 0. \qquad (29)$$

So it is sufficient to consider just the last one of (28)

$$\frac{dE^*_{2k}}{dz} = \frac{j\omega_2}{2}\sqrt{\frac{\mu_0}{\varepsilon_2}}\sum d_{ijk}E_{1i}E^*_{3j}e^{-j(k_1-k_3+k_2)z} + c.c.$$

$$\frac{dE_{3j}}{dz} = -j\frac{\omega_3}{2}\sqrt{\frac{\mu_0}{\varepsilon_3}}\sum d_{ijk}E_{1i}E_{2k}e^{-j(k_1+k_2-k_3)z} + c.c.. \qquad (28)$$

$$\frac{dE_{3j}}{dz} = -j\omega\sqrt{\frac{\mu_0}{\varepsilon}}\sum d_{jik}E_{1i}E_{1k}e^{j\Delta k\cdot z} \qquad (30)$$

where $\omega = \omega_1 = \frac{\omega_3}{2}$

and $\Delta k = k_3^{(j)} - k_1^{(i)} - k_1^{(k)}. \qquad (31)$

In eq.(31) $k_1^{(i)}$ is the constant of propagation of the beam at ω_1 polarized in the direction i. The solution of (30) for $E_{3j}(0) = 0$ for a crystal of length L is

$$E_{3j}(L) = -j\omega\sqrt{\frac{\mu_0}{\varepsilon}}\sum d_{jik}E_{1i}E_{1k}\frac{e^{j\Delta k\cdot L}-1}{j\Delta k}$$

or

$$I(L) = \left|E_{3j}(L)\right|^2 = \frac{\mu_0}{\varepsilon}\omega^2\left|\sum d_{jik}E_{1i}E_{1k}\right|^2 L^2\frac{\operatorname{sen}^2(\Delta k\cdot L/2)}{(\Delta k\cdot L/2)^2}. \qquad (32)$$

According to (32) a requirement for an efficient second harmonic generation is that $\Delta k = 0$, that is from (31) with $\omega_3 = 2\omega$, $\omega_1 = \omega_2 = \omega$

$$\Delta k = k_3^{(j)} - k_1^{(i)} - k_1^{(k)}. \qquad (31)$$

$$k^{(2\omega)} = 2k^{(\omega)}. \qquad (33)$$

If $\Delta k \neq 0$, the second harmonic wave generated at a generic plane z_1 which propagates until another plane z_2 is not in phase with that generated in z_2. This produces an interference described by the factor

$$\frac{\mathrm{sen}^2\left(\Delta k \cdot L/2\right)}{\left(\Delta k \cdot L/2\right)^2}$$

in (32).

$$I(L) = \left|E_{3j}(L)\right|^2 = \frac{\mu_0}{\varepsilon}\omega^2 \left|\sum d_{jik} E_{1i} E_{1k}\right|^2 L^2 \frac{\mathrm{sen}^2\left(\Delta k \cdot L/2\right)}{\left(\Delta k \cdot L/2\right)^2}. \qquad (32)$$

The condition (33) is never practically satisfied because, due to dispersion, the refractive index depends on ω.

$$k^{(2\omega)} = 2k^{(\omega)}. \qquad (33)$$

Therefore, we have

$$\Delta k = k^{(2\omega)} - 2k^{(\omega)} = \frac{2\omega}{c}\left(n^{(2\omega)} - n^{(\omega)}\right) \qquad (34)$$

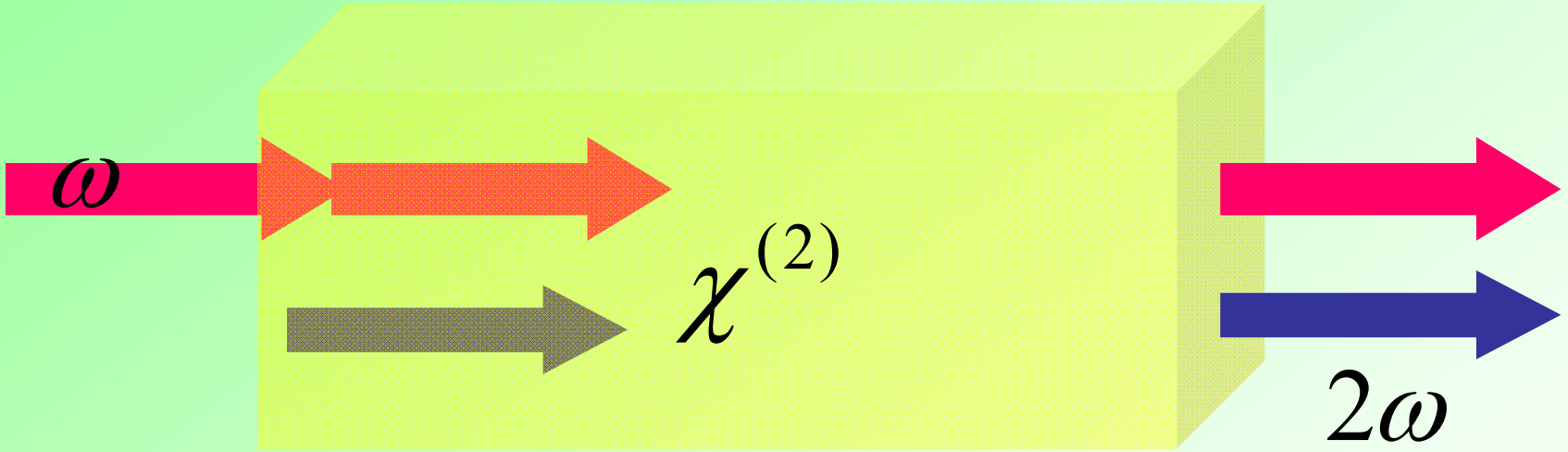
being

$$k^{(\omega)} = \frac{\omega n^{(\omega)}}{c} \qquad (35)$$

and therefore

$$\Delta k \neq 0.$$

Phase Matching



- Since the optical (NLO) media are dispersive, The fundamental and the harmonic signals have different propagation speeds inside the media.

- The harmonic signals generated at different points interfere destructively with each other.

Coherence length

We have no more second harmonic production when

$$\mathrm{Sin}(\Delta kL/2)/\Delta kL/2 = 0$$

This is achieved when $\Delta kL/2 = \pi$

Which means $L = 2\pi/\Delta k$

that is named coherence length

However, it is possible to make $\Delta k = 0$ (**phase-matching condition**) using various skills; the most used of which takes advantage from the natural birefringence of the anisotropic crystals. From (34) we can see that $\Delta k = 0$ implies

$$n^{(2\omega)} \simeq n^{(\omega)} \qquad (36)$$

$$\Delta k = k^{(2\omega)} - 2k^{(\omega)} = \frac{2\omega}{c}\left(n^{(2\omega)} - n^{(\omega)}\right) \qquad (34)$$

so that the refractive indices of second harmonic and of fundamental frequency have to be equal.

In the materials with normal dispersion, the index of the ordinary and extraordinary wave along a direction increase with ω, as it is shown in the table.

λ, μm	Index	
	n_o (ordinary beam)	n_s (extraordinary beam)
0,2000	1,622630	1,563913
0,3000	1,545570	1,498153
0,4000	1,524481	1,480244
0,5000	1,5144928	1,472486
0,6000	1,509274	1,468267
0,7000	1,505235	1,465601
0,8000	1,501924	1,463708
0,9000	1,498930	1,462234
1,0000	1,496044	1,460993
1,1000	1,493147	1,459884
1,2000	1,490169	1,458845
1,3000	1,487064	1,457838
1,4000	1,483803	1,456838
1,5000	1,480363	1,455829
1,6000	1,476729	1,454797
1,7000	1,472890	1,453735
1,8000	1,468834	1,452636
1,9000	1,464555	1,451495
2,0000	1,460044	1,450308

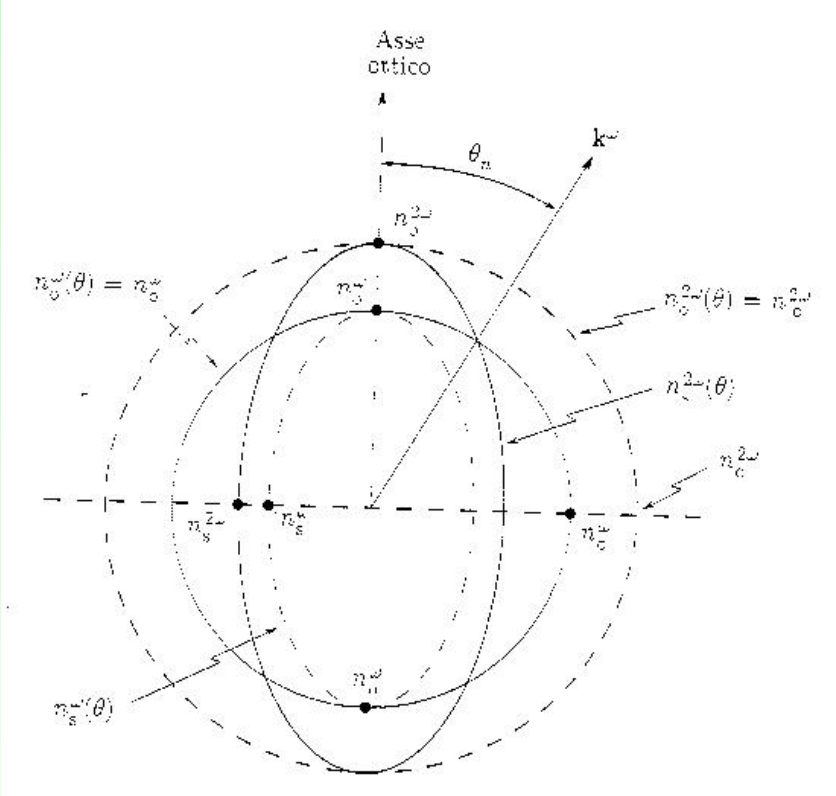
This makes it possible to satisfy eq.(36) when both the beams are of the same kind (that is both extraordinary or ordinary). Or (36) can be satisfied, in some cases, using an ordinary and an extraordinary wave.

$$n^{(2\omega)} \simeq n^{(\omega)} \qquad (36)$$

In order to illustrate this point we can consider the dependence of the refractive index of the extraordinary wave in a uniaxial crystal, from the angle ϑ between the direction of propagation and the optical axis (z) of the crystal.

$$\frac{1}{n_s^2(\vartheta)} = \frac{\cos^2\vartheta}{n_0^2} + \frac{\mathrm{sen}^2\vartheta}{n_s^2}. \qquad (37)$$

If $n_s^{(2\omega)} < n_0^{(\omega)}$ an angle ϑ_n exists for which $n_s^{(2\omega)}(\vartheta_n) = n_0^{(\omega)}$. In this case if the fundamental beam (frequency ω) is propagated along ϑ_n as a ordinary beam, the second harmonic beam will be generated along the same direction as an extraordinary beam. This situation is shown in the figure.



The angle ϑ_n is determined by the intersection between the sphere (shown as a circle in the figure) which corresponds to the index surface of the ordinary beam to ω with the index ellipsoid of the extraordinary beam. The angle ϑ_n, for negative uniaxial crystals (that is for crystals for which $n_s^{(2\omega)} < n_0^{(\omega)}$ is given by

$$\frac{\cos^2 \vartheta_n}{\left[n_0^{(2\omega)}\right]^2} + \frac{\operatorname{sen}^2 \vartheta_n}{\left[n_s^{(2\omega)}\right]^2} = \frac{1}{\left[n_0^{(\omega)}\right]^2} \qquad (38)$$

that is

$$\operatorname{sen}^2 \vartheta = \frac{\left[n_0^{(\omega)}\right]^{-2} - \left[n_0^{(2\omega)}\right]^{-2}}{\left[n_s^{(2\omega)}\right]^{-2} - \left[n_0^{(2\omega)}\right]^{-2}}. \qquad (39)$$

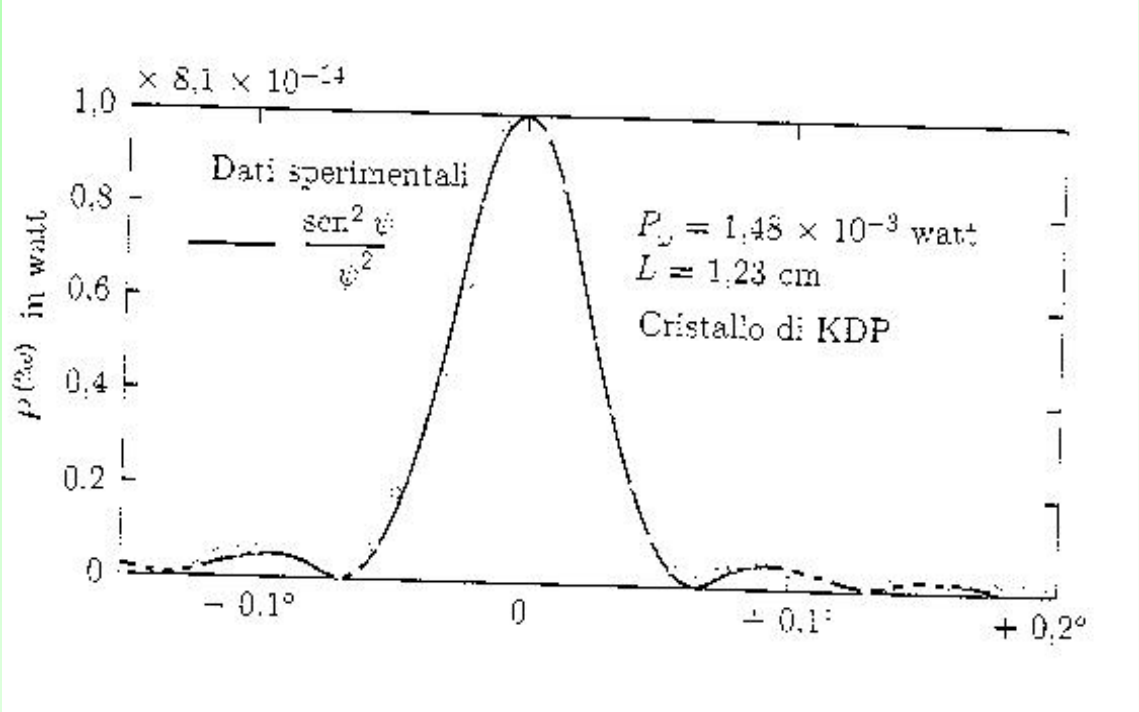
According to (32), if we deviate from the matching condition, for a fixed length L of the nonlinear crystal, we have a reduction of the second harmonic power generated by the factor

$$\frac{P^{(2\omega)}}{P_{max}^{(2\omega)}} = \frac{\text{sen}^2\left(\Delta k \cdot L/2\right)}{\left(\Delta k \cdot L/2\right)^2}. \qquad (40)$$

$$I(L) = \left|E_{3j}(L)\right|^2 = \frac{\mu_0}{\varepsilon}\omega^2 \left|\sum d_{jik} E_{1i} E_{1k}\right|^2 L^2 \frac{\text{sen}^2\left(\Delta k \cdot L/2\right)}{\left(\Delta k \cdot L/2\right)^2}. \qquad (32)$$

This relation can be easily verified varying the angle $\sigma = \vartheta - \vartheta_n$ between the direction of index matching and the propagation direction.

A diagram of the second harmonic power according to σ is shown in the figure (where the theoretical curve $(\text{sen } x/x)^2$ is also shown).



In the case of nanostructures, phase matching is not so important because in any case the radiation propagates over lengths which are comparable with the wavelength and so decoherence is negligible

Optical conservation laws

Conservation of momentum

$k(2\omega) = 2k(\omega)$

Conservation of energy

$2\omega = \omega + \omega$

$2\hbar\omega = \hbar\omega + \hbar\omega$

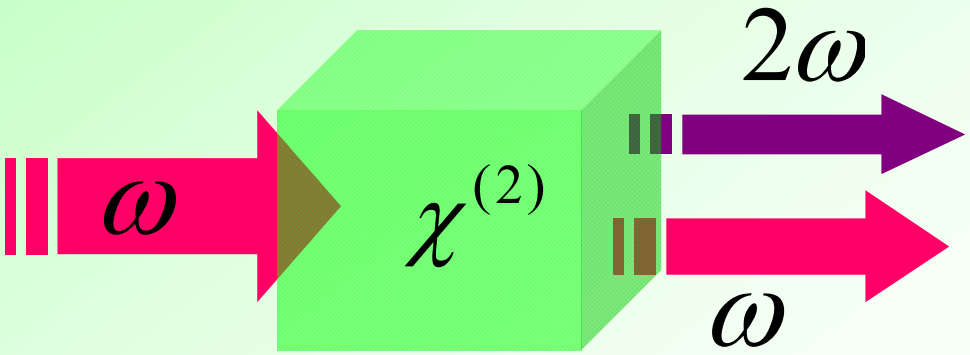
Nonlinear Optical Interactions

- The E-field of a laser beam

$$\tilde{E}(t) = Ee^{-i\omega t} + \text{C.C.}$$

- 2nd order nonlinear polarization

$$\tilde{P}^{(2)}(t) = 2\chi^{(2)}EE^{*} + (\chi^{(2)}E^{2}e^{-2i\omega t} + \text{C.C.})$$



2nd Order Nonlinearities

- The incident optical field

$$\tilde{E}(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + C.C.$$

- Nonlinear polarization contains the following terms

$$P(2\omega_1) = \chi^{(2)} E_1^2 \quad \text{(SHG)}$$

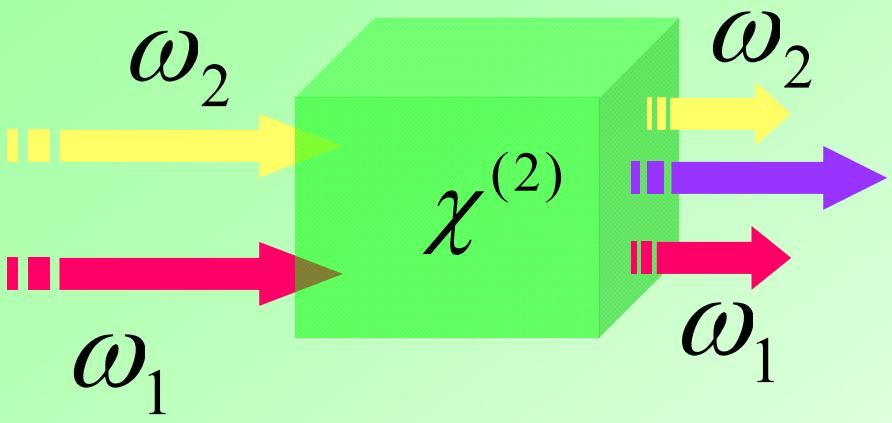
$$P(2\omega_2) = \chi^{(2)} E_2^2 \quad \text{(SHG)}$$

$$P(\omega_1 + \omega_2) = 2\chi^{(2)} E_1 E_2 \quad \text{(SFG)} \quad \text{summ frequency generation}$$

$$P(\omega_1 - \omega_2) = 2\chi^{(2)} E_1 E_2^* \quad \text{(DFG)} \quad \text{difference frequency generation}$$

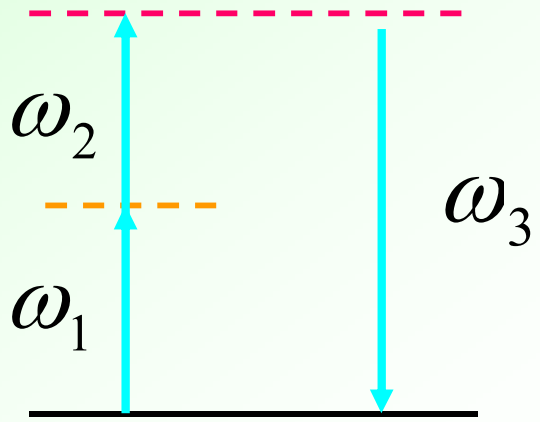
$$P(0) = 2\chi^{(2)} (E_1 E_1^* + E_2 E_2^*) \quad \text{(OR)}$$

Sum Frequency Generation

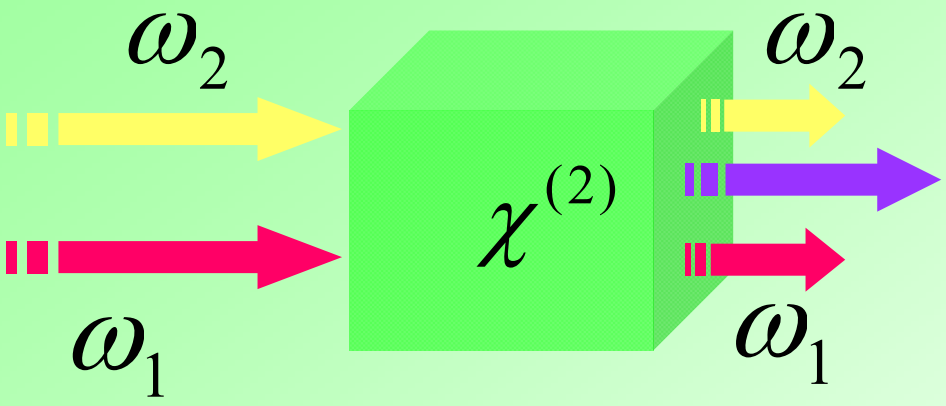


$$\omega_3 = \omega_1 + \omega_2$$

Application:
Tunable radiation in the UV Spectral region.

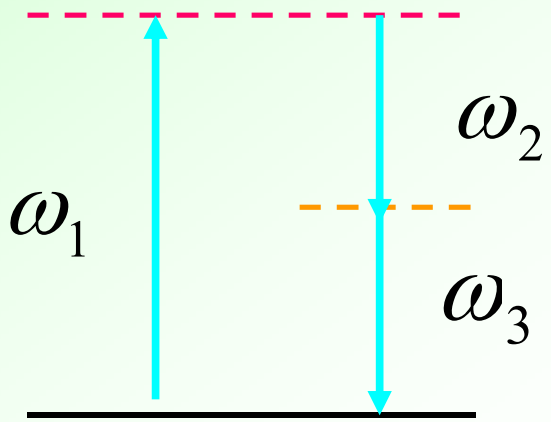


Difference Frequency Generation



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

Application:
The low frequency photon, ω_2 amplifies in the presence of high frequency beam ω_1 . This is known as parametric amplification.



Third Order Nonlinearities

When the general form of the incident electric field is in the following form,

$$\tilde{E}(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + E_3 e^{-i\omega_3 t}$$

The third order polarization will have 22 components

$$\omega_i, 3\omega_i, (\omega_i + \omega_j + \omega_k), (\omega_i + \omega_j - \omega_k)$$
$$(2\omega_i + \omega_j), (2\omega_i - \omega_j), i, j, k = 1,2,3$$

The Intensity Dependent Refractive Index

- The incident optical field

$$\tilde{E}(t) = E(\omega)e^{-i\omega t} + \text{C.C}$$

- Third order nonlinear polarization

$$P^{(3)}(\omega) = 3\chi^{(3)}(\omega = \omega + \omega - \omega)\,|E(\omega)|^2\,E(\omega)$$

The total polarization can be written as

$$P^{TOT}(\omega)=\chi^{(1)}E(\omega)+3\chi^{(3)}(\omega=\omega+\omega-\omega)|E(\omega)|^2 E(\omega)$$

One can define an effective susceptibility

$$\chi_{\text{eff}}=\chi^{(1)}+4\pi|E(\omega)|^2\ \chi^{(3)}$$

The refractive index can be defined as usual

$$n^2=1+4\pi\chi_{\text{eff}}$$

By definition

$$n = n_0 + n_2 I$$

where

$$I = \frac{n_0 c}{2\pi} |E(\omega)|^2$$

$$n_2 = \frac{12\pi^2}{n_0^2 c} \chi^{(3)}$$

Typical values of nonlinear refractive index

Mechanism	n_2 (cm^2/W)	(esu)	Response time (sec)
Electronic Polarization	10^{-16}	10^{-14}	10^{-15}
Molecular Orientation	10^{-14}	10^{-12}	10^{-12}
Electrostriction	10^{-14}	10^{-12}	10^{-9}
Saturated Atomic Absorption	10^{-10}	10^{-8}	10^{-8}
Thermal effects	10^{-6}	10^{-4}	10^{-3}
Photorefractive Effect	large	large	Intensity dependent

Third order nonlinear susceptibility of some material

Material	$\chi^{(3)}$	Response time
Air	1.2×10^{-17}	
CO_2	1.9×10^{-12}	2 Ps
GaAs (bulk room temperature)	6.5×10^{-4}	20 ns
CdS_xSe_{1-x} doped glass	10^{-8}	30 ps
GaAs/GaAlAs (MQW)	0.04	20 ns
Optical glass	$(1\text{-}100) \times 10^{-14}$	Very fast