The Abdus Salam
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Preparatory School

to the

Winter College on Fibre Optics, Fibre Lasers and Sensors

5 - 9 February 2007

Introduction to Non Linear Optics

Session 1

M.Centini
Universita' di Roma, "La Sapienza"
Roma, Italy

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Introduction to Non Linear Optics
1st Session

M.Centini
Universita' di Roma, "La Sapienza", Roma, Italy

Outline

1st session

Introduction:
-The Non linear Optical Susceptibility Tensor;

-On the Physical Origins of the Nonlinear Optical Coefficients;

Second Order Optical Nonlinearities:
-Second Harmonic Generation, Phase matching, examples of phase matching techniques;

-Basic equations of Parametric Amplification, Parametric Oscillation, three-wave mixing, frequency up- and down-conversion.

-**Third Order Optical Nonlinearities:**
-The Nonlinear Constants, Intensity dependence of the refractive index;

-Third harmonic generation; Third order parametric processes, four wave mixing;

-Self Focusing of Optical Beams, Diffraction-less propagation of Optical beams: Spatial Solitons;

2nd session

Third Order Optical Nonlinearities:
-Molecular Raman Scattering,

-Stimulated Raman Scattering;

- Stimulated Brillouin Scattering.

Introduction to Optical Solitons:
- Wave packets, Group velocity and Dispersion;

- Nonlinear wave packets and the nonlinear Schrödinger equation;

- Self phase modulation, Spectral broadening,

- Effects on group velocity dispersion, Pulse Compression;

- Modulation instability;

- Fundamental and higher orders Solitons.

Introduction to non linear optics

DEFINITION: **Non linear optics is the study of how high intensity light interacts with and propagates through matter.**

Before the discovery of the Laser, optics seemed to be a **linear** science. Nonlinear effects were hidden because of the relatively low intensities that occurred naturally or could be attained in the laboratory.

Sun light: |E| is of the order of 600 V/m; **Binding fields of the H atom: 10^{11} V/m**

What are the words "linear" and "nonlinear" referred to?

If we apply an optical field to a material system, the response of the material can be expressed in terms of an induced polarization, proportional to the strength of the applied field: (**local response**)

$$\vec{P}(\vec{r},t) = \varepsilon_0 \int_{-\infty}^{t} \hat{\chi}^{(1)}(t,t')\vec{E}(\vec{r},t')dt'$$

As a first approximation, the polarization is a linear function of the applied field.

Introduction to non linear optics

Superposition principle is still valid: For a given applied field, if we consider the Fourier transform:

$$\tilde{\vec{E}}(\vec{r},\omega) = \int_{-\infty}^{+\infty} \vec{E}(\vec{r},t)e^{i\omega t}dt$$

The FT of the Polarization is:

$$\tilde{\vec{P}}(\vec{r},\omega) = \varepsilon_0 \hat{\chi}^{(1)}(\omega)\tilde{\vec{E}}(\vec{r},\omega)$$

Thus:

$$\vec{P}(\vec{r},t) = \frac{1}{2\pi}\int_{-\infty}^{+\infty} \tilde{\vec{P}}(\vec{r},\omega)e^{-i\omega t}d\omega = \frac{1}{2\pi}\int_{-\infty}^{+\infty} \varepsilon_0 \hat{\chi}^{(1)}(\omega)\tilde{\vec{E}}(\vec{r},\omega)e^{-i\omega t}d\omega$$

Introduction to non linear optics

For a given input field:

$$\vec{E}(\vec{r},t) = \sum_{j=1,N} \vec{\xi}_j(\vec{r}) e^{-i\omega_j t}$$

$$\vec{P}(\vec{r},t) = \varepsilon_0 \sum_{j=1,N} \hat{\chi}^{(1)}(\omega_j)\vec{\xi}_j(\vec{r}) e^{-i\omega_j t} = \sum_{j=1,N} \vec{p}_j(\vec{r}) e^{-i\omega_j t}$$

For a superposition of fields with several angular frequencies, the induced polarization is a linear superposition of the polarization induced by each monochromatic component.

There is not the creation of new frequencies, Polarization vector oscillates with the same frequency components of the optical fields that induced it.

Introduction to non linear optics

Nonlinear contribution

Nevertheless, the medium response function is, in general, a complex function of the optical field. When the strength of the applied field is comparable to the atomic field strength, the linear approximation is no longer valid. By performing a power expansion of P it is possible to consider the **nonlinear** terms in the polarization function:

$$\vec{P}(t) = \vec{P}^{(1)}(t) + \vec{P}^{(2)}(t) + \vec{P}^{(3)}(t) + \ldots + \vec{P}^{(n)}(t)$$

Where:

$$\vec{P}^{(n)}(t) = \varepsilon_0 \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \ldots \int_{-\infty}^{t_{n-1}} dt_n \cdot \hat{\chi}^{(n)}(t, t_1, t_2, \ldots, t_n) \vdots \vec{E}(t_1)\vec{E}(t_2)\ldots\vec{E}(t_n)$$

Nonlinear terms in the polarization vector are responsible for coupling of fields at different frequencies and for creation of new frequencies.

Introduction to non linear optics

Lasers made available highly coherent radiation that could be concentrated and focused to give extremely high local intensities that can reach 10^{18} W/cm^2.

In 1961 Bloembergen's group started a program in a field that became known as "Nonlinear Optics". The early results are incorporated in a monograph of this title, published by W. A. Benjamin, New York, in 1965

Experimental observation of second harmonic generation by Franken et al. in 1961 considerably increased the interest of the community in the field of nonlinear optics.

A rich stream of new phenomena soon followed. Nonlinear optics plays an important role in telecommunications and future computer technologies. The relatively long interaction lengths and small cross sections available in waveguides and fibers means that low energy optical pulses can achieve sufficiently high peak intensities to put in evidence non linear effects also in many transparent optical materials with weak nonlinearities.

The Non linear Optical Susceptibility Tensor

Maxwell equations:

$$\nabla\times\vec{E}=-\frac{\partial\vec{B}}{\partial t},$$
$$\nabla\times\vec{H}=\frac{\partial\vec{D}}{\partial t}+\vec{j},$$
$$\nabla\cdot\vec{D}=\rho,$$
$$\nabla\cdot\vec{B}=0.$$

Must be supplemented by relations that describe the behavior of materials under the influence of the fields. (constitutive relations) that are generally very complicated.

If the e.m field is time-harmonic and if the bodies are at rest or in very slow motion relative to each other:

$$\vec{j}=\sigma\vec{E},\quad \vec{D}=\varepsilon_0\hat{\varepsilon}_r\vec{E},\quad \vec{B}=\mu_0\hat{\mu}_r\vec{H}.$$

Insulators or dielectrics: **σ (conductivity) is negligibly small; μ_r=1;**
thus we will focus on the electric properties of the materials.

An intense quasi monochromatic light pulse can, either directly or indirectly, excite material oscillation modes covering an enormous frequency bandwidth ranging from 10^{15} s^{-1} (electronic oscillations) to 1 s^{-1} (thermal oscillations).

The Non linear Optical Susceptibility Tensor

A proper theoretical treatment of these many diverse linear and nonlinear material oscillations must include a proper description of the polarization vector induced by the electric field. These quantities are linked through Maxwell's equations via the constitutive relation that in the general case can be written as:

$$\vec{D}(\vec{r},t)=\varepsilon_0\vec{E}(\vec{r},t)+\vec{P}(\vec{r},t)$$

Vacuum contribution

Material contribution

The Polarization vector is a complex function of the electric field. **For time-harmonic fields, homogeneous media and considering instantaneous material response to the electric field we can write:**

$$\vec{P}=\varepsilon_0\hat{\chi}^{(1)}\vec{E}+\varepsilon_0\hat{\chi}^{(2)}:\vec{E}\vec{E}+\varepsilon_0\hat{\chi}^{(3)}\vdots\vec{E}\vec{E}\vec{E}+\ldots=\vec{P}^{(1)}+\vec{P}^{(2)}+\vec{P}^{(3)}+\ldots$$

A) $\chi^{(n)}$**=nth order susceptibility. Optical properties depend on light intensity although large electric fields are needed.**

B) $\chi^{(2)}, \chi^{(4)},\ldots, \chi^{(2n)}=0$ **in materials with inversion symmetry(centrosymmetric) i.e. all isotropic materials (gases, liquids, glasses) and some type of crystals (NaCl for example)**

The Non linear Optical Susceptibility Tensor

Linear susceptibility tensor:

$$\vec{P}^{(1)}=\varepsilon_0\hat{\chi}^{(1)}\vec{E}$$

For anisotropic media $\hat{\chi}^{(1)}$ is a second rank tensor with 9 components (3x3 matrix).

For isotropic media

$$\hat{\chi}^{(1)}=\chi^{(1)}\begin{pmatrix}1&0&0\\0&1&0\\0&0&1\end{pmatrix}\longrightarrow\boxed{\vec{P}^{(1)}=\varepsilon_0\chi^{(1)}\vec{E}}$$

Nonlinear susceptibility tensor:

$$P_i^{(2)}=\varepsilon_0\sum_{j,k=x,y,z}\hat{\chi}_{ijk}^{(2)}E_jE_k;\quad i=x,y,z$$

$\hat{\chi}^{(2)}$ Third rank tensor (27 components) [m/V]

$$P_i^{(3)}=\varepsilon_0\sum_{j,k,l=x,y,z}\hat{\chi}_{ijkl}^{(3)}E_jE_kE_l;\quad i=x,y,z$$

$\hat{\chi}^{(3)}$ Fourth rank tensor (81 components) [m^2/V^2]

The Non linear Optical Susceptibility Tensor

Only non centrosymmetric crystals can posses a non-vanishing second order nonlinear susceptibility tensor.

In a centrosymmetric crystal, for every point (x, y, z) in the unit cell there is an indistinguishable point (-x, -y, -z). Thus a reversal of the sign of E_j and E_k must cause a reversal in the sign of $P_i^{(2)}$:

$$P_i^{(2)}\left(-E_j,-E_k\right)=\varepsilon_0\sum_{j,k=x,y,z}\hat{\chi}_{ijk}^{(2)}(-E_j)(-E_k)=-P_i^{(2)}\left(E_j,E_k\right)$$

Possible only if all the elements of the nonlinear susceptibility tensor are zero.

If the non linear susceptibility is independent of frequency, no physical significance can be attached to an exchange of E_j and E_k. Thus

$$\hat{\chi}_{ijk}^{(2)}=\hat{\chi}_{ikj}^{(2)}$$

18 physically distinct components remain. The nonlinear optical coefficient tensor is defined as follows:

The Non linear Optical Susceptibility Tensor

$$\begin{pmatrix}P_x^{(2)}\\P_y^{(2)}\\P_z^{(2)}\end{pmatrix}=2\varepsilon_0\begin{pmatrix}d_{11}&d_{12}&d_{13}&d_{14}&d_{15}&d_{16}\\d_{21}&d_{22}&d_{23}&d_{24}&d_{25}&d_{26}\\d_{31}&d_{32}&d_{33}&d_{34}&d_{35}&d_{36}\end{pmatrix}\begin{pmatrix}E_xE_x\\E_yE_y\\E_zE_z\\2E_yE_z\\2E_zE_x\\2E_xE_y\end{pmatrix}$$

Where:

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

i=1,2,3,=x,y,z
l=1=xx; l=2=yy; l=3=zz;
l=4=yz=zy; l=5=zx=xz;
l=6=xy=yx.

It can be shown that if the material is transparent (lossless) in the frequency range containing all the frequencies involved in the nonlinear process (Kleinman symmetry):

$$\hat{\chi}_{ijk}^{(2)}=\hat{\chi}_{ikj}^{(2)}=\hat{\chi}_{kij}^{(2)}=\hat{\chi}_{kji}^{(2)}=\hat{\chi}_{jik}^{(2)}=\hat{\chi}_{jki}^{(2)}$$

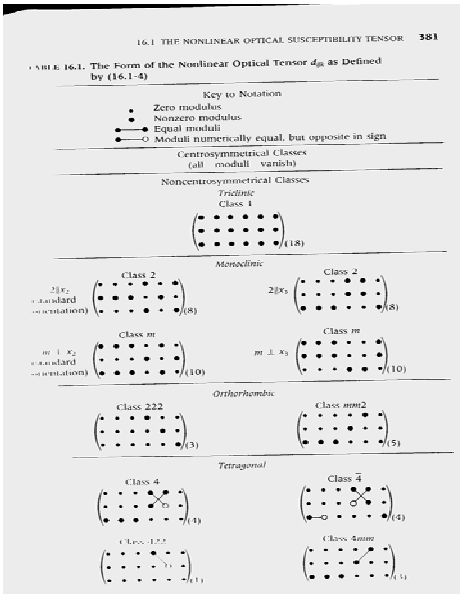
The maximum number of independent d coefficients is reduced to 10:

$$\begin{pmatrix}d_{11}&d_{12}&d_{13}&d_{14}&d_{15}&d_{16}\\d_{16}&d_{22}&d_{23}&d_{24}&d_{14}&d_{12}\\d_{15}&d_{24}&d_{33}&d_{23}&d_{13}&d_{14}\end{pmatrix}$$

The non zero elements are related to the point group symmetry of the crystal.

The Non linear Optical Susceptibility Tensor

A. Yariv, Quantum electronics



16.1 THE NONLINEAR OPTICAL SUSCEPTIBILITY TENSOR 381

TABLE 16.1. The Form of the Nonlinear Optical Tensor d_{jk} as Defined by (16.1-4)

Key to Notation
Zero modulus
Nonzero modulus
Equal moduli
Moduli numerically equal, but opposite in sign

Centrosymmetrical Classes (all moduli vanish)

Noncentrosymmetrical Classes

Triclinic
Class 1 (18)

Monoclinic
Class 2 (8)
Class m (10)

Orthorhombic
Class 222 (3)
Class mm2 (5)

Tetragonal
Class 4 (1)
Class $\bar{4}$ (4)

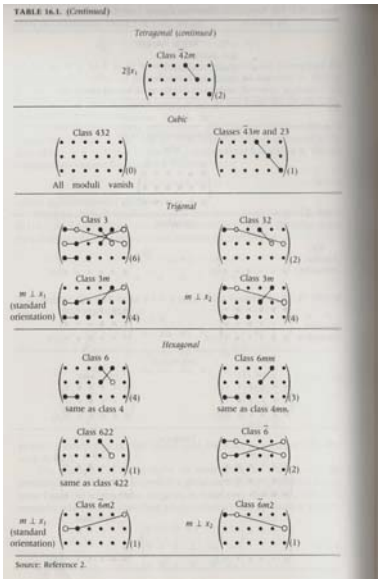


TABLE 16.1. (Continued)

Tetragonal (continued)
Class $\bar{4}2m$ (2)

Cubic
Class 432 (0) — All moduli vanish
Classes $\bar{4}3m$ and 23 (1)

Trigonal
Class 3 (6)
Class 32 (2)
Class 3m (4) — $m \perp x_1$ (standard orientation)
Class 3m (4) — $m \perp x_1$

Hexagonal
Class 6 (4) — same as class 4
Class 6mm (3) — same as class 4mm
Class 622 (1) — same as class 422
Class $\bar{6}$ (2)
Class $\bar{6}m2$ (1) — $m \perp x_1$ (standard orientation)
Class $\bar{6}m2$ (1) — $m \perp x_2$

Source: Reference 2.

The Non linear Optical Susceptibility Tensor

TABLE 1.5.3 Second-order nonlinear optical susceptibilities for several crystals

Material	Point group	d_{ij} (10^{-9} esu)
Quartz	$32 = D_3$	$d_{11} = 0.96$ $d_{14} = 0.02$
$Ba_2NaNb_5O_{15}$	$mm2 = C_{2v}$	$d_{31} = -35$ $d_{32} = -35$ $d_{33} = -48$
$LiNbO_3$	$3m = C_{3v}$	$d_{22} = 7.4$ $d_{31} = 14$ $d_{33} = 98$
$BaTiO_3$	$4mm = C_{4v}$	$d_{15} = -41$ $d_{31} = -43$ $d_{33} = -16$
$NH_4H_2PO_4$ (ADP)	$\bar{4}2m = D_{2d}$	$d_{14} = 1.2$ $d_{36} = 1.2$
KH_2PO_4 (KDP)	$\bar{4}2m = D_{2d}$	$d_{14} = 1.2$ $d_{36} = 1.1$
$LiIO_3$	$6 = C_6$	$d_{35} = -13$ $d_{36} = -10$
CdSe	$6mm = C_{6v}$	$d_{15} = 74$ $d_{31} = 68$ $d_{33} = 130$
KD_2PO_4 (KD*P)	$\bar{4}2m = D_{2d}$	$d_{36} = 1.26$ $d_{14} = 1.26$
CdS	$6mm = C_{6v}$	$d_{33} = 86$ $d_{31} = 90$ $d_{36} = 100$
Ag_3AsS_3 (proustite)	$3m = C_{3v}$	$d_{22} = 68$ $d_{31} = 36$
$CdGeAs_2$	$\bar{4}2m = D_{2d}$	$d_{36} = 1090$
$AgGaSe_2$	$\bar{4}2m = D_{2d}$	$d_{36} = 81$
$AgSbS_3$ (pyrargyrite)	$3m = C_{3v}$	$d_{31} = 30$ $d_{22} = 32$
β-BaB_2O_4 (beta barium borate)		$d_{11} = 4.6$

To obtain the value of d in units of [m/V], multiply each entry by: $4\pi/(3x10^4)=4.189x10^{-4}$

For example the KH_2PO_4 (KDP) has a $D_{2d}=\bar{4}2m$ point group symmetry:

$d_{14}=(1.26x10^{-9})(4.189x10^{-4})=$
$=5.28x10^{-13}$ m/V = 0.5 pm/V

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

Exercise: Show that for a laser beam propagating along the KDP optic axis (E_z=0), the nonlinear $\mathbf{P}^{(2)}$ vector points along the optical axis, irrespective of the polarization of the incident laser beam.

On the Physical Origins of the Nonlinear Optical Coefficients;

We focus our attention to the electronic response to a driving electric field.
Non linear behavior can be due to a non resonant response or to a resonant response:

Non resonant non linear response

Laser is tuned to the transparency region of a crystal;
Far from resonance with any atomic transitions.

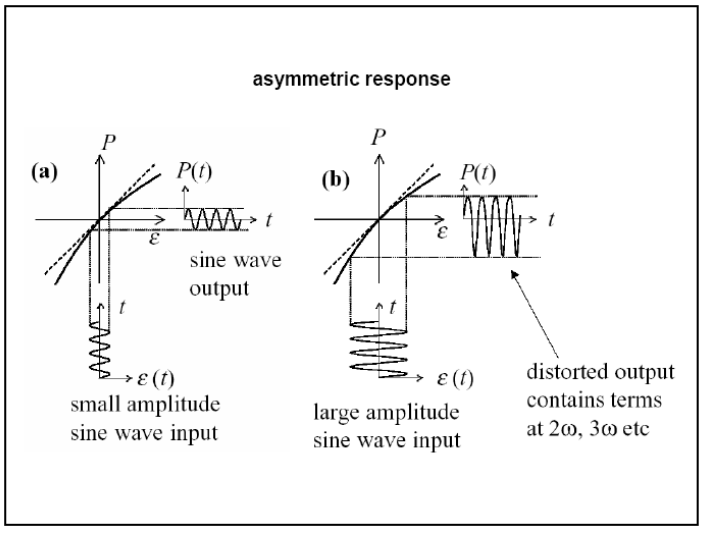
The model assumes that an electron is bound to the nucleus by an anharmonic potential. The equation of motion for the electron is:

$$\ddot{X} + \gamma\dot{X} + \omega_0^2 X + DX^2 = \frac{eE_0}{2m}\left(e^{i\omega t} + e^{-i\omega t}\right)$$

Where: **X** is the displacement with respect to the equilibrium position
γ is the damping term
$E_0\cos(\omega t)$ is the driving electric field
$-mDX^2$ is the anharmonic restoring force (corresponding to (m/3)DX^3 potential)

On the Physical Origins of the Nonlinear Optical Coefficients;

The anharmonic response causes a distortion in the dynamic because the electron experiences a stronger force for +X than –X.



On the Physical Origins of the Nonlinear Optical Coefficients;

We assume the solution has the form:

$$X(t) = \frac{1}{2}\left(X_1 e^{i\omega t} + X_2 e^{i2\omega t} + c.c.\right), \quad X_1 >> X_2$$

Substituting to the equation we found the values of X_1 and X_2. Then:

$$P(\omega,t) = -NeX(\omega,t) = \varepsilon_0 \chi^{(1)}(\omega)E(t);$$
$$P(2\omega,t) = -NeX(2\omega,t) = \varepsilon_0 \chi^{(2)}(\omega)E(t)^2;$$

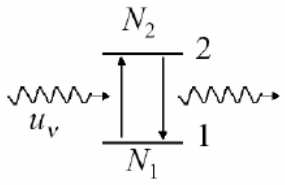
And finally:

$$\chi^{(1)}(\omega) = \frac{Ne^2}{m\varepsilon_0\left(\omega_0^2 - \omega^2 + i\gamma\omega\right)};$$
$$\chi^{(2)}(\omega) = \frac{mD\chi^{(1)}(\omega)^2\chi^{(1)}(2\omega)\varepsilon_0^2}{N^2e^3};$$

Exercise: Derive the expressions for the linear and the non linear susceptibilities solving the system of equations for *P(ω,t)* and *P(2ω,t).*

On the Physical Origins of the Nonlinear Optical Coefficients;

Resonant non linear response



- laser is tuned to resonance with an atomic transition
- stimulated emission reduces net absorption rate at high intensity
- equivalent to "Pauli blocking" of upper state
- saturable absorption: (empirically found)
  $\alpha(I) = \alpha_0 / (1 + I / I_s)$
  I_s = saturation intensity, depends on the transition rate
- $\alpha(I) \propto \alpha_0 - \alpha_2 I$ at small I
- $\alpha \propto \mathrm{Im}(\varepsilon_r)$, $I \propto E^2 \Rightarrow \varepsilon_r \propto E^2$, ie., third-order nonlinearity

$$\vec{P} = \varepsilon_0\left(\chi^{(1)} + \beta\left|E\right|^2\right)\vec{E}$$

Second order nonlinear effects

We start with Maxwell equations in a form, which includes the polarization vector:

(a) $$\nabla\times\vec{E}=-\frac{\partial\left(\mu_0\vec{H}\right)}{\partial t},$$

(b) $$\nabla\times\vec{H}=\frac{\partial\vec{D}}{\partial t}+\vec{j}=\vec{j}+\frac{\partial}{\partial t}\left(\varepsilon_0\vec{E}+\vec{P}\right),$$

We consider time harmonic fields and we expand the polarization vector up to the second order:

$$\vec{P}=\vec{P}^{(1)}+\vec{P}^{(2)}=\varepsilon_0\hat{\chi}^{(1)}\vec{E}+\varepsilon_0\hat{\chi}^{(2)}:\vec{E}\vec{E}$$

Equation (b) can be written as:

$$\nabla\times\vec{H}=\sigma\vec{E}+\frac{\partial}{\partial t}\varepsilon_0\varepsilon_r\vec{E}+\frac{\partial}{\partial t}\left(\varepsilon_0\hat{\chi}^{(2)}:\vec{E}\vec{E}\right),$$

After taking the curl of both sides of the equation (a) and replacing the expression (c) reminding that:

$$\nabla\times\nabla\times\vec{E}=\nabla\nabla\cdot\vec{E}-\nabla^2\vec{E};$$

$$\nabla\cdot\vec{E}=0;\quad\text{(Homogenous non charged medium)}$$

Second order nonlinear effects

$$\nabla^2\vec{E}=\mu_0\sigma\frac{\partial}{\partial t}\vec{E}+\mu_0\varepsilon_0\varepsilon_r\frac{\partial^2}{\partial t^2}\vec{E}+\mu_0\varepsilon_0\frac{\partial^2}{\partial t^2}\left(\hat{\chi}^{(2)}:\vec{E}\vec{E}\right),$$

At this point we specialize the problem to one dimension by taking $\partial/\partial x=\partial/\partial y=0$
We denote the arbitrary direction of propagation as z.

We also limit the consideration to three frequencies

$$E_i^{(\omega_1)}(z,t)=\frac{1}{2}\left[E_{1i}(z)e^{i(\omega_1 t-k_1 z)}+c.c.\right]$$
$$E_k^{(\omega_2)}(z,t)=\frac{1}{2}\left[E_{2k}(z)e^{i(\omega_2 t-k_2 z)}+c.c.\right]$$
$$E_j^{(\omega_3)}(z,t)=\frac{1}{2}\left[E_{3j}(z)e^{i(\omega_3 t-k_3 z)}+c.c.\right]$$

Where *i,j,k* refer to the Cartesian coordinates and can each take on values x and y.
Note that in the linear case the solution is given by the same expression where the envelopes are independent of z.

Second order nonlinear effects

Why are we limiting the study to three frequencies and what are the implications?

If we considered only one monochromatic field, the non linear polarization:

$$\vec{P}^{(2)} = \varepsilon_0 \hat{\chi}'^{(2)} : \vec{E}\vec{E} = \varepsilon_0 \hat{\chi}'^{(2)} : \left(\frac{1}{2}\left(E_1(z)e^{i(\omega_1 t - k_1 z)} + c.c\right)\right)\left(\frac{1}{2}\left(E_1(z)e^{i(\omega_1 t - k_1 z)} + c.c\right)\right)$$

Contains terms oscillating at $\omega_1+\omega_1=2\omega_1$, $\omega_1-\omega_1=0$

Thus the non linear polarization acts as a source for e.m. field at different frequencies. Description of the electric field as a single frequency field is no longer adequate.

In a more general case we consider that the electric field contains three frequencies fulfilling the requirement $\omega_1=\omega_3-\omega_2$. **(energy conservation requirement)**

$$\vec{P}^{(2)} = \varepsilon_0 \hat{\chi}'^{(2)} : \vec{E}\vec{E} = \varepsilon_0 \hat{\chi}'^{(2)} : \left(\vec{E}^{(\omega_1)} + \vec{E}^{(\omega_2)} + \vec{E}^{(\omega_3)}\right)\left(\vec{E}^{(\omega_1)} + \vec{E}^{(\omega_2)} + \vec{E}^{(\omega_3)}\right)$$

NOTE: **$\chi'^{(2)}$ is the tensor $\chi^{(2)}$ transformed from the crystal coordinate system to that used here to describe the field propagation**

Second order nonlinear effects

Performing the products it is evident that polarization vector contains terms oscillating at several frequencies i.e.

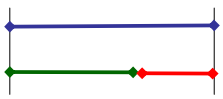
$2\omega_1$, $\omega_1+\omega_2=\omega_3$, $\omega_1+\omega_3$, $\omega_2-\omega_1$, $\omega_3-\omega_1=\omega_2$, $2\omega_2$, $\omega_2+\omega_3$, $\omega_1-\omega_2$, $\omega_3-\omega_2=\omega_1$, 0, $2\omega_3$, $\omega_1-\omega_3$, $\omega_2-\omega_3$.

We assume only a finite number of frequencies is involved in the process, by neglecting creation of new frequencies. Thus we consider only the polarization terms oscillating at frequencies ω1 ω2 and ω3.

From a practical point of view, it is possible to find conditions such that the other processes involving different frequencies have negligible efficiencies with respect to the one involving frequencies ω_1 ω_2 and ω_3. (Phase matching)

The non linear polarization at ω_3 acts as a source which eventually creates a photon of energy $\hbar\omega_3$ by destroying two photons, one of energy $\hbar\omega_1$ and the other of energy $\hbar\omega_2$. In the process energy is conserved if:

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



In this case the equation can be separated in three coupled equations, one for each frequency component.

Second order nonlinear effects

Back to our equation:

$$\nabla^2\vec{E} = \mu_0\sigma\frac{\partial}{\partial t}\vec{E} + \mu_0\varepsilon_0\varepsilon_r\frac{\partial^2}{\partial t^2}\vec{E} + \mu_0\varepsilon_0\frac{\partial^2}{\partial t^2}\left(\hat{\chi}^{(2)} : \vec{E}\vec{E}\right),$$

In our one dimensional model we have:

$$\nabla^2\vec{E}(z,t) = \frac{\partial^2}{\partial z^2}\left(\vec{E}_{(\omega_1)}(z,t) + \vec{E}_{(\omega_2)}(z,t) + \vec{E}_{(\omega_3)}(z,t)\right),$$

Being *i,j,k* the Cartesian coordinates and can each take on values x and y.

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

$$E_k^{(\omega_2)}(z,t) = \frac{1}{2}\left[E_{2k}(z)e^{i(\omega_2 t - k_2 z)} + c.c.\right]$$

$$E_j^{(\omega_3)}(z,t) = \frac{1}{2}\left[E_{3j}(z)e^{i(\omega_3 t - k_3 z)} + c.c.\right]$$

Performing the second order derivative and assuming that the variation of the complex field envelopes with z are small enough so that (nonlinearity is a small perturbation of the linear solution):

$$\frac{d}{dz}E_{1i}(z)k_1 >> \frac{d^2}{dz^2}E_{1i}(z),$$

Slow varying envelope approximation SVEA

Second order nonlinear effects

The evaluation of the Laplacian within the SVEA approximation gives:

$$\nabla^2\vec{E} = \frac{d^2}{dz^2}\left[\vec{E}^{(\omega_1)}(z,t) + \vec{E}^{(\omega_2)}(z,t) + \vec{E}^{(\omega_3)}(z,t)\right] \cong -\frac{1}{2}\left\{\begin{array}{l}\left[k_1^2E_{1i}(z) + 2ik_1\frac{dE_{1i}(z)}{dz}\right]e^{i(\omega_1 t - k_1 z)} + \\ + \left[k_2^2E_{2k}(z) + 2ik_2\frac{dE_{2k}(z)}{dz}\right]e^{i(\omega_2 t - k_2 z)} + \\ + \left[k_3^2E_{3j}(z) + 2ik_3\frac{dE_{3j}(z)}{dz}\right]e^{i(\omega_3 t - k_3 z)}\end{array}\right\}$$

Finally, with some algebra: [**Exercise:** derive the set of equations]

$$\frac{dE_{1i}}{dz} = -\frac{\sigma_1}{2}\sqrt{\frac{\mu_0}{\varepsilon_0\varepsilon_{r1}}}E_{1i} - \frac{i\omega_1}{2}\sqrt{\frac{\mu_0\varepsilon_0}{\varepsilon_{r1}}}\left(\sum_{j,k=x,y}\hat{\chi}'^{(2)}_{ijk}E_{3j}E^*_{2k}\right)e^{-i(k_3-k_2-k_1)z}$$

$$\frac{dE^*_{2k}}{dz} = -\frac{\sigma_2}{2}\sqrt{\frac{\mu_0}{\varepsilon_0\varepsilon_{r2}}}E^*_{2k} + \frac{i\omega_2}{2}\sqrt{\frac{\mu_0\varepsilon_0}{\varepsilon_{r2}}}\left(\sum_{i,j=x,y}\hat{\chi}'^{(2)}_{kij}E_{1i}E^*_{3j}\right)e^{-i(k_1-k_3+k_2)z}$$

$$\frac{dE_{3j}}{dz} = -\frac{\sigma_3}{2}\sqrt{\frac{\mu_0}{\varepsilon_0\varepsilon_{r3}}}E_{3j} - \frac{i\omega_3}{2}\sqrt{\frac{\mu_0\varepsilon_0}{\varepsilon_{r3}}}\left(\sum_{i,k=x,y}\hat{\chi}'^{(2)}_{jik}E_{1i}E_{2k}\right)e^{-i(k_1+k_2-k_3)z}$$

Second order nonlinear effects

NOTE: The non linear polarization for each frequency components can act as a gain, a loss or a phase modulation, depending on the modulus and the relative phase between the fields.

In order to have efficient energy transfer among the fields, a long interaction length is required, the phase mismatch should be as close as possible to zero. In other words:

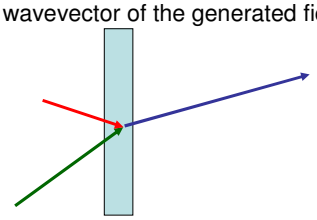
$$\mathbf{k_3=k_2+k_1}$$

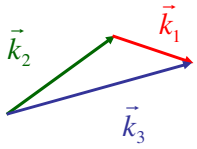
In a more general case, the condition to be fulfilled is:

$$\vec{k}_3 = \vec{k}_1 + \vec{k}_2$$

It is called phase matching condition and it can be interpreted as a momentum conservation requirement.

Example: Two pump fields at frequencies ω_1 and ω_2 can generate a sum frequency (ω_3) field. The wavevector of the generated field will fulfill:





Optical Second harmonic Generation

First experimental report on second harmonic generation was performed by Franken, Hill, Peters and Weinreich in 1961. (Fig: A. Yariv, Quantum electronics)

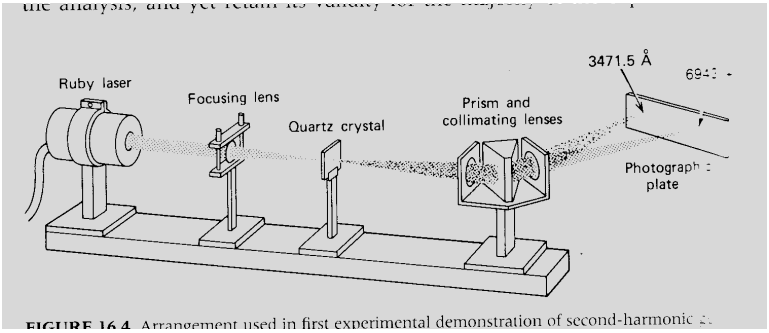


FIGURE 16.4 Arrangement used in first experimental demonstration of second-harmonic

SHG can be studied as the limiting case of the three frequency interaction where two of the frequencies ω_1 and ω_2 are equal and $\omega_3=2\omega_1$.

We assume as first approximation that the amount of power lost by the input beam is negligible so that $dE_{1i}/dz=0$ and the medium is transparent at ω_3 ($\sigma=0$)

Optical Second harmonic Generation

$$\frac{dE_{3j}}{dz} = -i\omega\sqrt{\frac{\mu_0\varepsilon_0}{\varepsilon_{r3}}}\left(\sum_{i,k=x,y}\hat{\chi}'^{(2)}_{jik}E_{1i}E_{1k}\right)e^{i\Delta kz}$$

Where: $\omega = \omega_1 = \omega_3/2$

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

$k_1^{(i)}$ is the propagation constant for the beam at ω_1 which is polarized along the i direction. The solution for $E_{3j}(0)=0$ (no SH input) and for a crystal of length L is:

$$E_{3j}(L) = -i\omega\sqrt{\frac{\mu_0\varepsilon_0}{\varepsilon_{r3}}}\left(\sum_{i,k=x,y}\hat{\chi}'^{(2)}_{jik}E_{1i}E_{1k}\right)\frac{e^{i\Delta kL}-1}{i\Delta k};$$

We note that:

$$\frac{e^{i\Delta kL}-1}{i\Delta k} = -ie^{i\frac{\Delta kL}{2}}\frac{L}{2}\frac{\left(e^{i\frac{\Delta kL}{2}}-e^{-i\frac{\Delta kL}{2}}\right)}{\Delta k\frac{L}{2}} = -ie^{i\frac{\Delta kL}{2}}\frac{L}{2}\frac{\sin\left(\Delta k\frac{L}{2}\right)}{\Delta k\frac{L}{2}};$$

Optical Second harmonic Generation

To obtain an expression for the second harmonic power output, considering A linearly polarized pump with non vanishing electric field component E_{1x} we use the relation:

$$\frac{P^{(2\omega)}}{Area} = \frac{1}{2}\varepsilon_0 c\sqrt{\varepsilon_{r3}}E_{3j}E_{3j}^*$$

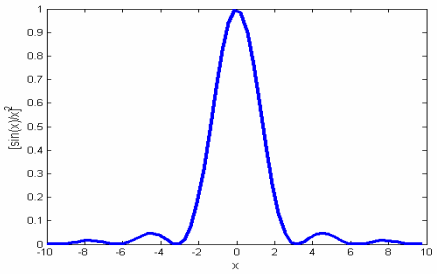
If we consider the SH generated linearly polarized with non vanishing field component E_{3y}we can write an expression for the conversion efficiency:

$$\eta = \frac{P^{(2\omega)}}{P^{(\omega)}} = \frac{1}{2}\frac{\omega^2L^2\left(\chi'^{(2)}_{yxx}\right)^2}{\varepsilon_0c^3n_{2\omega}n_\omega^2}\frac{P^{(\omega)}}{Area}\left[\frac{\sin(\Delta kL/2)}{\Delta kL/2}\right]^2;$$

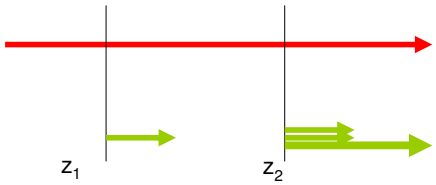
The conversion efficiency is linearly growing with the pump intensity. This means that the generated second harmonic intensity with respect to the pump intensity follows a quadratic law. The conversion efficiency increases with the squared length of the nonlinear medium

Phase matching

We note that the conversion efficiency is crucially determined by the sinc function:



Maximum conversion efficiency is achieved for ΔkL/2=0,
$\Delta k=k_{2\omega}-2k_{\omega}=0$ is called phase matching condition and it is fulfilled as long as
The refractive indices at FF and SH frequencies are the same: **$n_{2\omega}=n_{\omega}$.**
If such condition is fulfilled, the FF and SH fields propagate with the same phase velocities



Phase matching

In common materials the refractive index is an increasing function of the frequency thus we have $n_{\omega}<n_{2\omega}$, and **$\Delta k>0$**.

During the non linear process there is an exchange of energy between FF and SH field and the SH production is zero for any propagation length that satisfy the law:

$\Delta kL/2=m\pi$, where m is an integer.

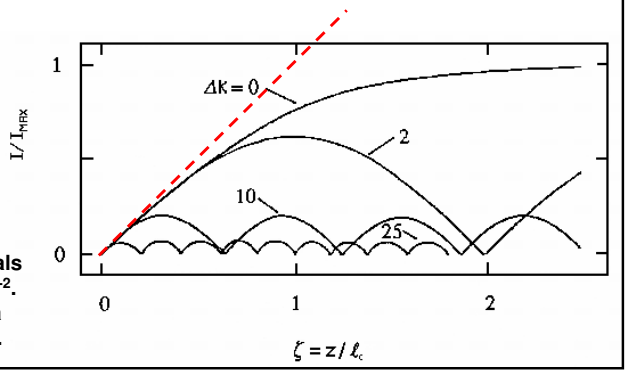
For a mismatched interaction, the length of non linear material that produce the maximum generated second harmonic field can be calculated by:

Undepleted pump approximation

$$\frac{\Delta k l_c}{2}=\frac{\pi}{2};$$

$$l_c=\frac{\pi}{\Delta k}=\frac{\lambda_0}{4(n_{2\omega}-n_{\omega})}$$

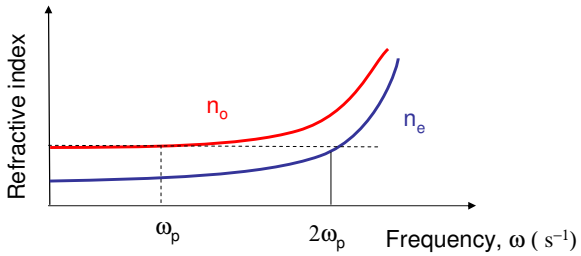
Typically in nonlinear materials Δn is of the order of $10^{-1}$to 10^{-2}. Coherence lengths are only a few numbers of wavelenghts.



Phase matching techniques

A widely used technique takes advantage of the natural birefringence of anisotropic crystals.

Under certain circumstances it is possible to use the different refractive indices for the ordinary wave and the extraordinary wave. For example, a typical behaviour of dispersion of the refractive indices of a negative ($n_e<n_o$) uniaxial crystal is:



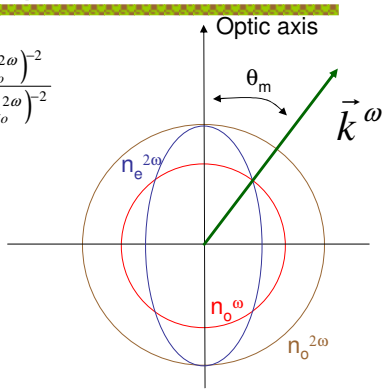
If $n_e(2\omega_p)<n_o(\omega_p)$, it exists an angle θ_m at which $n_e^{2\omega}(\theta_m)=n_o^{\omega}$. It can be calculated by:

$$\frac{1}{n_e^2(\theta)}=\frac{\cos^2\theta}{n_o^2}+\frac{\sin^2\theta}{n_e^2}$$

Phase matching techniques

TYPE I phase matching
o+o → e (neg. uniaxial)

$$\sin^2\theta_m=\frac{\left(n_o^{\omega}\right)^{-2}-\left(n_o^{2\omega}\right)^{-2}}{\left(n_e^{2\omega}\right)^{-2}-\left(n_o^{2\omega}\right)^{-2}}$$



Exercise: Ruby laser λ=694.3 nm on a KDP Crystal as an ordinary wave.
$n_e^{\omega}=1.466$; $n_e^{2\omega}=1.487$;
$n_o^{\omega}=1.506$; $n_o^{2\omega}=1.534$;
Calculate the matching angle

TYPE II phase matching
e+o → e (neg uniaxial)

Phase matching condition Δk=0 becomes:

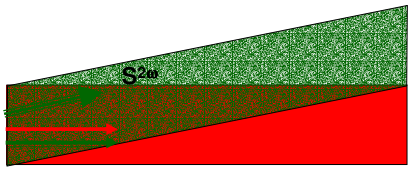
$$n_{je}^{2\omega}(\theta)=\frac{1}{2}\left[n_{io}^{\omega}+n_{ke}^{\omega}(\theta)\right]$$

Exercise:Calculate the index matching angle for type II second harmonic generation, show that:

$$\left[\frac{\cos^2\theta_m}{\left(n_o^{2\omega}\right)^2}+\frac{\sin^2\theta_m}{\left(n_e^{2\omega}\right)^2}\right]^{-1/2}=\frac{1}{2}\left\{n_o^{\omega}+\left[\frac{\cos^2\theta_m}{\left(n_o^{\omega}\right)^2}+\frac{\sin^2\theta_m}{\left(n_e^{\omega}\right)^2}\right]^{-1/2}\right\}$$

Phase matching techniques

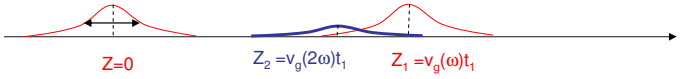
NOTE: Whenever the angle between the optic axis and the propagation direction has a value other than 0 and 90 degrees, the Poynting vector **S** and the propagation vector **k** are not parallel for extraordinary rays.
WALK OFF ANGLE limits the effective interaction length.



Similar argumentations apply if we consider second harmonic generation with ultra short pump pulses. Although phase velocities are matched, the group velocity is defined as:

$$v_g = \frac{c}{[n(\omega) + \omega(dn/d\omega)]}$$

Usually, $v_g(2\omega)$ is not the same as $v_g(\omega)$. This means that the pump wave packet will travel at a different velocity with respect to the generated second harmonic wave packet. The interaction between them stops when they are spatially separated.



$L_{MAX}=T_o/|d_{12}|$; where d_{12}=walk off parameter=$[v_g(\omega)]^{-1}$- $[v_g(2\omega)]^{-1}$

Exercise: derive the expression for L_{MAX}

Phase matching techniques

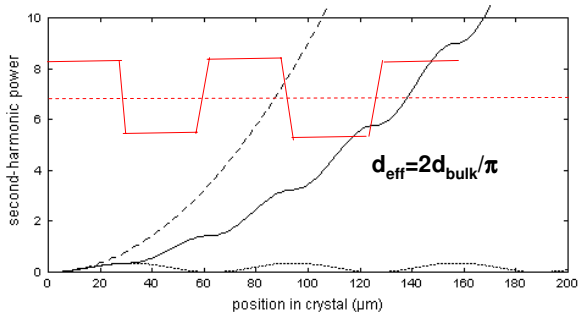
Quasi Phase matching

Periodic modulation of the non linear coefficients tensor elements responsible for the interaction.
It can be shown that the phase matching condition becomes:

$\Delta k=2\pi m/\Lambda$ Where m is an integer and Λ is the period of the nonlinearity.

EXAMPLE: If the sign of the non linear interaction is reversed at every coherence length d(z) is a periodic function of period ***$2l_c=2\pi/\Delta k$.***

QPM is achieved for m=1.



Phase matching techniques

Examples of applications of QPM

Periodically poled $LiNbO_3$
Periodical reversal of static electric field in order to induce a permanent periodically modulated electric polarization.

Fiber gratings:
The grating is prepared by sending intense FF and SH in the Fiber. Because of a third order process, a static DC polarization is created. Because of refractive index dispersion, the static field is spatially modulated, indeed:

$$E_{static} \propto E_{FF} E_{FF} E_{SH}^{*} + c.c. \propto E_0 e^{i\Delta k_p z} + c.c$$
$$\Delta k_p = \frac{2\omega_p}{c}\left[n_{2\omega_p} - n_{\omega_p}\right]$$

A periodic array of dipoles is created generating an effective second order susceptibility :

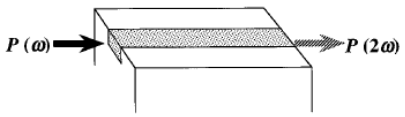
$$\hat{\chi}^{(2)}(z) = \hat{\chi}_0^{(2)} \cos(\Delta k_p z);$$

Then, if we send a pump at ω_p, we have QPM generation of SH

Phase matching techniques

Considering dielectric waveguides, the modal dispersion can be used to achieve phase matching. The phase matching relation for second harmonic generation becomes:

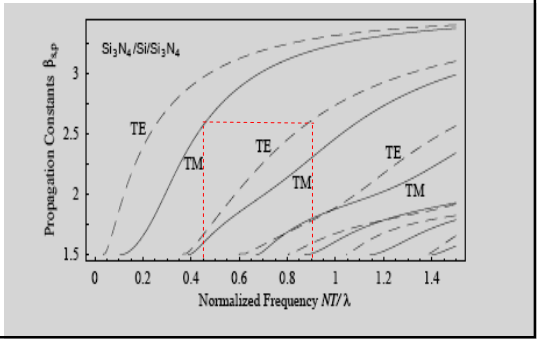
$$\tilde{\beta}_{2\omega} = \tilde{\beta}_{\omega}$$



This condition can be easily fulfilled by considering modes for the pump and for the SH of different order or polarization.

Nevertheless the **non linear process is governed by how the fields overlap: usually overlaps between fields belonging to different orders are not efficient.**

$$\eta \propto \left[\int_{-\infty}^{+\infty} \hat{\chi}^{(2)} : \left(\vec{E}_m^{\omega}(x)\right)^2 \cdot \left(\vec{E}_n^{2\omega}(x)\right)^{*} dx\right]^2$$

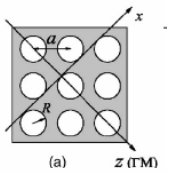


Phase matching techniques

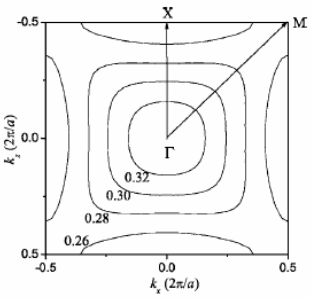
Photonic Crystals

In a photonic crystal **both the linear and the nonlinear susceptibility functions can be periodically modulated**. Modulation of the linear susceptibility is responsible for peculiar properties of the linear dispersion curves of these structures. Fields are characterized by a Bloch wave vector and an periodic function over the unit cell. Thus the quasi momentum conservation for second harmonic generat

$$\vec{k}_{2\omega} - \vec{k}_{\omega} - \vec{G} = \vec{0}$$



Where **G** is a reciprocal lattice vector. Usually unit cells are of the order of the wavelength or less unlike in the QPM regime where domains are inverted every coherence length.



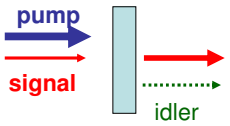
Optimum non linear interaction can be obtained by considering:

1- **distortion of the dispersion curves close to the band gaps. [phase matching, group velocity matching, density of states]**

2- **overlap of the fields with the chi(2) function over the unit cell. [enhancement of the effective non linear response, QPM]**

Basic Equations of Parametric Amplification

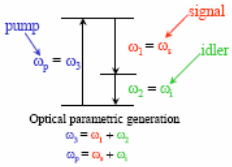
We consider an intense high frequency pump field (ω_3) and a lower frequency (ω_2) signal. Parametric process allows amplification of the signal. At the same time an idler field is generated at frequency $\omega_1=\omega_3-\omega_2$.



$$\frac{dE_1}{dz} = -\frac{i\omega_1}{2}\sqrt{\frac{\mu_0\varepsilon_0}{\varepsilon_{r1}}}\hat{\chi}'^{(2)}E_3E_2^*e^{-i\Delta kz}$$

$$\frac{dE_2^*}{dz} = +\frac{i\omega_2}{2}\sqrt{\frac{\mu_0\varepsilon_0}{\varepsilon_{r2}}}\hat{\chi}'^{(2)}E_1E_3^*e^{i\Delta kz}$$

$$\frac{dE_3}{dz} = -\frac{i\omega_3}{2}\sqrt{\frac{\mu_0\varepsilon_0}{\varepsilon_{r3}}}\hat{\chi}'^{(2)}E_1E_2e^{-i\Delta kz}$$



$\Delta k = k_3 - k_2 - k_1$

For simplicity we neglect absorption, and we only consider linearly polarized fields along the x direction. We choose the z axis as propagation direction. We also define:

$$A_l = \sqrt{\frac{n_l}{\omega_l}}E_l; \qquad l = 1,2,3. \quad \Longrightarrow \quad I_l = \frac{1}{2}c\varepsilon_0 n_l|E_l|^2 = \frac{1}{2}c\varepsilon_0\omega_l|A_l|^2$$

Since a photon's energy is $\hbar\omega_l$, $|A_l|^2$ is proportional to the photon flux at ω_l

Basic Equations of Parametric Amplification

$$\frac{dA_1}{dz} = -i\kappa A_3 A_2^* e^{-i\Delta kz}$$
$$\frac{dA_2^*}{dz} = +i\kappa A_1 A_3^* e^{i\Delta kz}$$
$$\frac{dA_3}{dz} = -i\kappa A_1 A_2 e^{-i\Delta kz}$$

where: $\kappa = \frac{\hat{\chi}'^{(2)}}{2c}\sqrt{\frac{\omega_1\omega_2\omega_3}{n_1 n_2 n_3}}$

Exercise: derive the set of equations starting from the previous one.

In the undepleted pump approximation we take $A_3(z)=A_3(0)$ and we integrate the first two equations:

$$\frac{dA_1}{dz} = -igA_2^* e^{-i\Delta kz}$$
$$\frac{dA_2^*}{dz} = +igA_1 e^{i\Delta kz}$$

where: $g = \kappa A_3(0)$

Solutions (Phase matching, Δk=0):

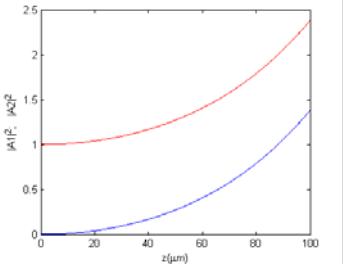
$$A_1(z) = A_1(0)\cosh(gz)$$
$$A_2(z) = iA_1(0)\sinh(gz)$$

NOTE: Beside the effect of parametric amplification of the signal beam we have generation of the idler beam. Since $\omega_2=\omega_3-\omega_1$, the process of generating a low frequency beam from two higher frequency fields is called **DIFFERENCE FREQUENCY GENERATION**

Basic Equations of Parametric Amplification

The increase in power of signal and idler waves Is at the expense of the pump wave.
It can be shown that neglecting absorption losses:

$$-\frac{d}{dz}\left(A_3 A_3^*\right) = \frac{d}{dz}\left(A_2 A_2^*\right) = \frac{d}{dz}\left(A_1 A_1^*\right)$$

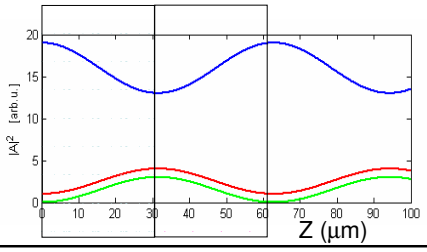


Manley-Rowe relation: It means that for each photon subtracted from the pump, a signal photon and an idler photon are created

Exercise: Consider a $LiNbO_3$ crystal pumped by a pump with λ_p=0.5 μm and I_p=5x10^6 W/cm^2 We use the d_{15} to amplify a signal at λ_s=1 μm. Refractive index at λ_s is 2.2. Calculate the gain available in the parametric process.

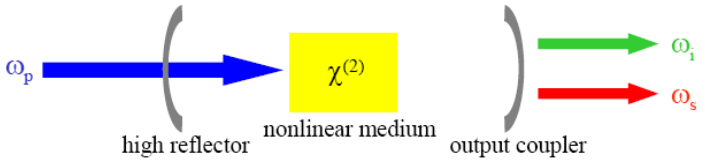
The general solutions, including the phase mismatch and depletion of the pump show a continuous exchange of energy between the fields. Depending on the initial condition:

Parametric amplification
Difference frequency generation
Down conversion
Sum frequency generation



Parametric Oscillation

Place mirrors reflective at ω_s and/or ω_i on either side of non linear medium in order to have an optical resonator.
Oscillation occurs as a consequence of the **gain** of the parametric amplification process. There is no need for the weak signal beam since it is generated internally to the crystal as so called "parametric noise".



For a given pump frequency, signal and idler frequencies are determined by:
1- energy conservation (frequency matching): $\omega_p=\omega_s+\omega_i$
2- Momentum conservation (Phase matching): $k_p=k_s+k_i$
Assuming the pump as an extraordinary wave we can tune the signal and idler frequencies rotating the crystal. following tuning condition:

$$\omega_3 n_{3e}(\theta) = \omega_1 n_{1o} + \omega_2 n_{2o}$$

OPO are widely used as coherent, tunable sources. In principle ω_s can be tuned in a range from 0 to ω_p. In real systems tunability depends on the birefringence and dispersion of the crystal and geometrical adaptability of the system.

Three wave mixing

Summary

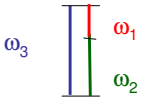
The basic equations govern the interaction of three fields due to second order nonlinearity:

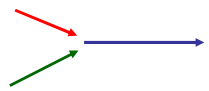
$$\frac{dE_1}{dz} = -\frac{i\omega_1}{2}\sqrt{\frac{\mu_0\varepsilon_0}{\varepsilon_{r1}}}\hat{\chi}'^{(2)}E_3E_2^*e^{-i\Delta kz}$$

$$\frac{dE_2^*}{dz} = +\frac{i\omega_2}{2}\sqrt{\frac{\mu_0\varepsilon_0}{\varepsilon_{r2}}}\hat{\chi}'^{(2)}E_1E_3^*e^{i\Delta kz}$$

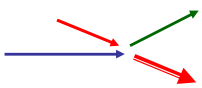
$$\frac{dE_3}{dz} = -\frac{i\omega_3}{2}\sqrt{\frac{\mu_0\varepsilon_0}{\varepsilon_{r3}}}\hat{\chi}'^{(2)}E_1E_2e^{-i\Delta kz}$$

Different processes can happen depending on the initial condition:

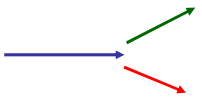




Sum frequency generation
Frequency up-conversion



Parametric amplification
Difference frequency generation
Stimulated parametric down conversion



Spontaneous parametric down conversion
Spontaneous parametric fluorescence

Third order optical nonlinearities

Third order non linear effects are present in every kind of material. The nonlinear polarization vector for **harmonic fields** **and instantaneous medium response** can be written as:

$$P_i^{(3)} = \varepsilon_0 \sum_{j,k,l=x,y,z} \hat{\chi}_{ijkl}^{(3)} E_j E_k E_l; \quad i = x, y, z$$

$\hat{\chi}^{(3)}$ Fourth rank tensor (81 components) [m^2/V^2]

For crystalline solids with low symmetry all 81 of these elements are independent and can be nonzero. The number of independent elements is reduced with the symmetry. **For isotropic materials there are only 3 independent elements.**

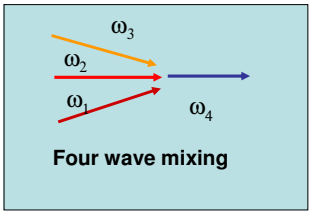
A- each of the coordinate axes must be equivalent:
B- nonzero elements must possess the property that any cartesian index that appears at least once, appears an even number of times.

$$\chi_{xxxx} = \chi_{yyyy} = \chi_{zzzz};$$
$$\chi_{xxyy} = \chi_{xxzz} = \chi_{yyxx} = \chi_{yyzz} = \chi_{zzxx} = \chi_{zzyy};$$
$$\chi_{xyxy} = \chi_{xzxz} = \chi_{yzyz} = \chi_{yxyx} = \chi_{zxzx} = \chi_{zyzy};$$
$$\chi_{xyyx} = \chi_{xzzx} = \chi_{yxxy} = \chi_{yzzy} = \chi_{zxxz} = \chi_{zyyz};$$

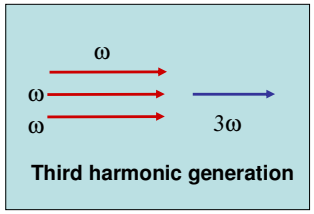
These elements are linked by the relation: $\chi_{xxxx} = \chi_{xxyy} + \chi_{xyxy} + \chi_{xyyx}$ derived from the requirement that the elements must be the same when calculated in two different coordinate systems that are rotated with respect to each other.

Third order optical nonlinearities

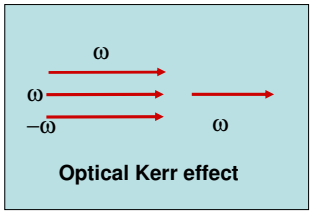
Third order nonlinear polarization allow coupling of fields at different frequencies, Here we present an sketched overview of the most relevant effects that will be analyzed later:



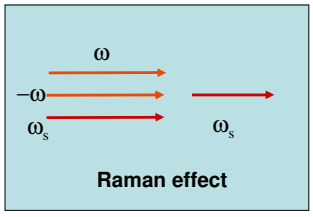
Four wave mixing



Third harmonic generation



Optical Kerr effect



Raman effect

Intensity dependence of the refractive index

The refractive index of many materials depends on the intensity of the incident optical field. If we consider an optical field of the form:

$$\vec{\mathrm{E}}(t)=\frac{1}{2}\left[E(\omega)e^{-i\omega t}+c.c\right]\hat{x}$$

Time averaged intensity is defined:

$$I=\frac{1}{2}c\varepsilon_0 n_0\left|E(\omega)\right|^2$$

The nonlinear refractive index is defined:

$$\tilde{n}\left(\omega,\left|E(\omega)\right|^2\right)=n_0(\omega)+n_2\left|E(\omega)\right|^2 \qquad \textbf{or} \qquad \tilde{n}\left(\omega,\left|E(\omega)\right|^2\right)=n_0(\omega)+n_{2I}I$$

Using the definition of P_{NL}:

$$\vec{\mathrm{P}}_{NL}=\varepsilon_0\hat{\chi}^{(3)}\vdots\vec{\mathrm{E}}\vec{\mathrm{E}}\vec{\mathrm{E}}$$

Intensity dependence of the refractive index

It contains terms oscillating at several frequencies. If we collect the terms oscillating at ω:

$$\mathrm{P}_{NL}(\omega)=\varepsilon_0\varepsilon_{NL}E(\omega)$$

where:

$$\varepsilon_{NL}=\frac{3}{4}\chi^{(3)}_{xxxx}\left|E(\omega)\right|^2$$

Exercise: **calculate the ε_{NL} by substituting the expression of the optical field into the nonlinear polarization.**

Reminding that: $\varepsilon_r=\left(\tilde{n}\right)^2=1+\chi^{(1)}(\omega)+\varepsilon_{NL}$

Using the relation: $\tilde{n}=n_0+n_2\,|\,E\,|^2=n_0\sqrt{1+\frac{\varepsilon_{NL}}{n_0^2}}\cong n_0\left(1+\frac{\varepsilon_{NL}}{2n_0^2}\right)$

We found that: $n_2(\omega)=\frac{3}{8n_0(\omega)}\chi^{(3)}_{xxxx}$ **or** $n_{2I}(\omega)=\frac{3}{4c\varepsilon_0 n_0^2(\omega)}\chi^{(3)}_{xxxx}$;

Intensity dependence of the refractive index

Boyd, Non linear Optics

TABLE 4.1.2 Third-order nonlinear susceptibilities of various materials*

Material	$\chi^{(3)}_{1111}$ (esu)	Response time
Air (20°C)	1.2×10^{-17}	
Carbon disulfide	1.9×10^{-12}	2 ps
GaAs (bulk, excitonic, room temperature)	6.5×10^{-4}	20 ns
GaAs/GaAlAs (MQW)	0.04	20 ns
Indium antimonide (77 K, 5.4 μm)	0.3	400 ns
Semiconductor-doped glass (containing CdSe)	10^{-8}	30 ps
Optical glasses	$(1-100) \times 10^{-14}$	Very fast
Polydiacetylene:		
Nonresonant	2.5×10^{-10}	Very fast
At peak of exciton	7.5×10^{-6}	2 ps

* The value of n_2 defined by $n = n_0 + n_2 I$ in units of cm^2/W can be obtained by multiplying the value of $\chi^{(3)}_{1111}$ by $0.0395/n_0^2$. The value of $\bar{n}_2$ defined by $n = n_0 + 2\bar{n}_2|E|^2$ in units of cm^2/erg can be obtained by multiplying the value of $\chi^{(3)}_{1111}$ by $3\pi/n_0$.

TABLE 4.1.1 Typical values of the nonlinear refractive index*

Mechanism	n_2 (cm^2/W)	$\chi^{(3)}_{1111}$ (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^{-9}	10^{-4}	10^{-3}
Photorefractive effect†	(large)	(large)	(intensity-dependent)

* For linearly polarized light, n_2 and $\chi^{(3)}$ are accurately related by Eq. (4.1.20).
† The photorefractive effect often leads to a very strong nonlinear response. This response usually cannot be described in terms of a $\chi^{(3)}$ (or an n_2) nonlinear susceptibility, because the nonlinear polarization does not depend on the applied field strength in the same manner as the other mechanisms listed.

Exercise: Calculate the value of Δn for carbon disulfide when it is applied an optical field of intensity 1 MW/cm^2. What is the Δn for optical glass in the same condition?

Intensity dependence of the refractive index

Intensity dependence of the refractive index has drastic effects both on the spatial properties of optical beams and on temporal (spectral) properties of ultrashort pulses.

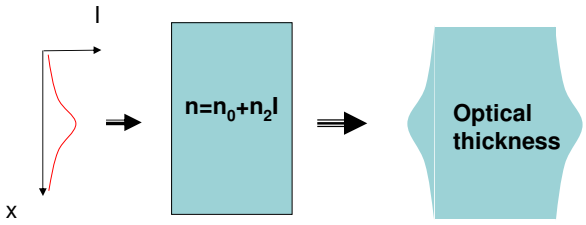
Self phase modulation (SPM)
Phase shift experienced by short pulse propagating through a nonlinear refractive index
- spectral broadening;
- Optical solitons in anomalous dispersion regimes of fibres.

Cross phase modulation (XPM)
Phase shift of a field induced by a co-propagating intense field at different wavelength.

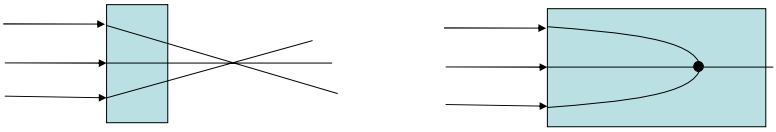
Self focusing and self defocusing of optical beams
Depending on the sign on the nonlinearity, the central (more intense) portion of a Beam experiences higher (lower) index of refraction with respect to the outer edges.An effective lens is created.

Self focusing of optical beams

If n_2 is positive, the refractive index of the material is larger at the center of the laser beam than at its periphery, with the result that the medium is in effect turned into a positive lens.



If the medium is short enough, this focus will occour outside of the medium, other way it will occur inside the medium leading to a extremely intense filament that will eventually damage the material.



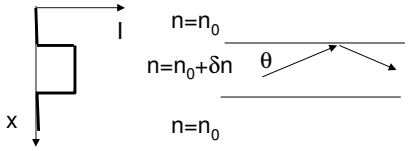
Self focusing of optical beams

Self-focusing can occur only if the power of the laser beam is sufficiently large. Other way diffraction effects will prevail and the beam will spread.

When the tendency of the beam to spread is precisely compensated by the self focusing effect, a phenomenon known as self trapping may occur. **Self trapping is usually unstable in 2D or 3D systems. Small perturbations in the beam diameter will lead either to rapid spreading of the beam or to catastrophic collapse.**

Total internal reflection occurs if $\theta<\theta_0$. where:

$$\cos\theta_0 = \frac{n_0}{n_0+\delta n} \cong 1-\frac{\delta n}{n_0}$$



A laser beam of diameter d will contain rays within a cone with an angle of the order of the characteristic diffraction angle:

$$\theta_d = \frac{0.61\lambda_0}{n_0 d}$$

Self focusing of optical beams

The diameter of the self trapped beam can be calculated by imposing $\theta_0 = \theta_d$

$$d = \frac{0.61\lambda_0}{\sqrt{2n_0 n_2 I}}$$

The power contained in a filament of diameter d is:

$$P_{cr} = \pi d^2 I = \frac{\pi(0.61\lambda_0)^2}{2n_0 n_2}$$

NOTE: The predicted power is independent of the beam diameter. Self focusing will occur if the power of the laser beam is greater than P_{cr}. Only if $P=P_{cr}$, self trapping is possible. For $P>P_{cr}$ the beam usually break up into several filaments, each of which contains power P_{cr}.

Exercise: For CS_2 (carbon disulfide), $n_2=2.6\times10^{-14}$ cm^2/W, $n_0=1.7$. Calculate P_{cr} at $\lambda_0=1$ μm
And for glasses? (see tables)

Spatial Solitons

Nonlinear Beam propagation equation:

$$\nabla\times\vec{E} = -\frac{\partial(\mu_0\vec{H})}{\partial t},$$
$$\nabla\times\vec{H} = \frac{\partial}{\partial t}\varepsilon_0\varepsilon_r\vec{E} + \frac{\partial}{\partial t}\left[\left(\varepsilon_{NL}\left\langle\vec{E}\cdot\vec{E}\right\rangle\right)\vec{E}\right],$$

Reminding the definition of ε_{NL} the wave equation becomes:

$$\nabla^2\vec{E} - \frac{n_0^2}{c^2}\frac{\partial^2}{\partial t^2}\vec{E} - \frac{2n_0 n_2}{c^2}\frac{\partial^2}{\partial t^2}\left[\left(\left\langle\vec{E}\cdot\vec{E}\right\rangle\right)\vec{E}\right] = 0,$$

We choose a field propagating along the z axis and polarized along the x direction:

$$\vec{E}(\vec{r},t) = \frac{1}{2}\left[\xi(\vec{r})e^{i(\omega t-kz)} + c.c.\right]\hat{x};$$
$$k = \frac{n_0\omega}{c};$$

Spatial Solitons

Performing the time averaged intensity, using SVEA we obtain the equation for the envelope functions: (Nonlinear Schrödinger Equation)

$$\nabla_T^2 \xi(\vec{r}) - 2ik\frac{\partial}{\partial z}\xi(\vec{r}) + \frac{n_2}{n}k^2\left|\xi(\vec{r})\right|^2\xi(\vec{r}) = 0,$$

Where

$$\nabla_T^2 = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2},$$

is the transverse Laplacian:

Exercise: Derive the expression for the non linear beam propagation equation

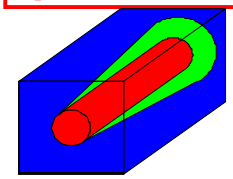
Spatial Solitons are special solution of the nonlinear Schrödinger equation.

1. **Robust balance between diffraction and a nonlinear beam narrowing process**
2. **Stationary solution to a nonlinear wave equation**
3. **Stable against perturbations**

Observed and Studied Experimentally to Date in:

1. **Kerr and saturating Kerr media**
2. **Photorefractive media**
3. **Quadratically nonlinear media**
4. **Liquid crystals**
5. **Gain media**

Spatial Solitons (2+1)D



Spatial Solitons

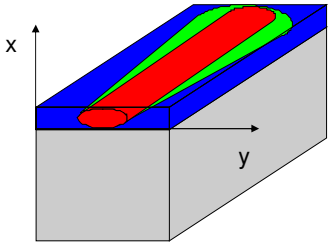
It can be shown that for NON SATURABLE KERR media, stable solutions exists only if the field is confined in one dimension. Otherway, we have the previously described phenomenon, diffraction less solutions exist only for a unique value of laser power. Higher powers leads to filamentation of the beam. At lower powers, diffraction wins.

Nevertheless it is possible to obtain **spatial solitons in (1+1)D using Kerr media and taking advantage of planar waveguide properties.**

For 1D confinement we have:

$$\frac{\partial^2}{\partial y^2}\xi(y,z) - 2ik\frac{\partial}{\partial z}\xi(y,z) + \frac{n_2}{n}k^2\left|\xi(y,z)\right|^2\xi(y,z) = 0,$$

Spatial Solitons (1+1)D



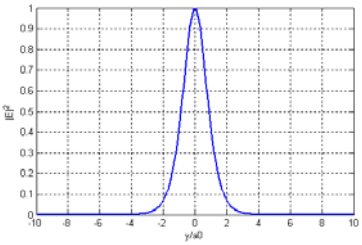
A requirement for being a soliton is that the modulus of the transverse profile does not change during propagation along z:

$$\frac{\partial}{\partial z}\left|\xi(y,z)\right| = 0,$$

Spatial Solitons

It is possible to show that the equation allows a family of solution fulfilling that requirement. These functions are called spatial solitons.
The fundamental soliton has the expression:

$$\xi(y,z)=\sqrt{\frac{n_0}{n_2}}\frac{1}{ka_0}\operatorname{sech}\left(\frac{y}{a_0}\right)e^{i\frac{z}{2ka_0^2}};$$



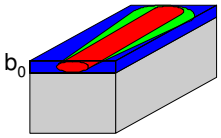
The integrated intensity over the beam is:

$$U_s=\int|\xi(y)|^2dy=\frac{n_0}{n_2}\frac{2}{k^2a_0};$$

The transverse mode size $\mathbf{b_0}$ will not be affected by the nonlinearity as long as $\mathbf{b_0<<a_0}$.

The Power required to launch a fundamental soliton is:

$$P=\frac{2n_0b_0}{k^2a_0n_2};$$



NOTE: The solution is stable, Pa_0=constant

Exercise 1(nonlinear susceptibility tensor): Show that for a laser beam propagating along the KDP optic axis (E_z=0), the nonlinear $P^{(2)}$ vector points along the optical axis, irrespective of the polarization of the incident laser beam.

Exercise 2 (Phase matching in birefringent crystals): Consider an intense ruby laser λ=694.3 nm on a KDP Crystal as an ordinary wave. Calculate the matching angle for SHG using the following data: n_e^{ω}=1.466; $n_e^{2\omega}$=1.487; n_o^{ω}=1.506; $n_o^{2\omega}$=1.534;

Exercise 3 (Parametric amplification): Consider a $LiNbO_3$ crystal pumped by a pump with λ_p=0.5 μm and $I_p=5\times10^6$ W/cm^2. We use the d_{15} to amplify a signal at λ_s=1 μm. Refractive index at λ_s is 2.2. Calculate the gain available in the parametric process.

Exercise 4 (Intensity dependent refractive index): Calculate the value of Δn for carbon disulfide when it is applied an optical field of intensity 1 MW/cm^2. What is the Δn for optical glass in the same condition?

Exercise 5 (Self focusing): For CS_2 (carbon disulfide), $n_2=2.6\times10^{-14}$ cm^2/W, n_0=1.7. Calculate P_{cr} for self trapping at λ_0=1 μm. And for glasses? (see tables)

Exercise 6 (Four wave mixing): Show why, in a normally dispersive and isotropic medium, the condition $2k_2=k_1+k_3$ (ω3>ω2>ω1; 2ω2=ω1+ω3) cannot be satisfied for propagation along a single direction.

Exercise 7 (Spatial solitons): Give an estimation of the power required to launch a 15 μm x 5 μm spot size soliton at λ_0 =0.62 μm in a glass waveguide.(n0=1.53; n2=3.4x10-16 cm^2/W); Give a physical interpretation to the statement made: "The transverse mode size b0 will not be affected by the nonlinearity as long as b0<<a0"