

## Unit III

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**Non-linear optics** :- It is study of phenomena that occur as a consequence of modification of optical properties of material by presence of light. The field of Nonlinear optics was began in 1961 when Franken discovered the phenomena of Second Harmonic generation. We know that polarization  $P$  of material depends on the strength of applied optical field  $E$  by following manner

$$P = \epsilon_0 \chi^{(1)} E(t)$$

Here  $\epsilon_0$  is permittivity of free space and  $\chi^{(1)}$  is known as linear susceptibility. In non linear optics the polarization  $P$  can be expressed as power series of  $E(t)$  as following way

$$P(t) = \epsilon_0 \left[ \chi^{(1)} E(t) + \chi^{(2)} E^2(t) + \chi^{(3)} E^3(t) + \dots \right] \quad \text{--- (1)}$$

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$$= P^{(1)}(t) + P^{(2)}(t) + P^{(3)}(t) + \dots$$

The quantity  $\chi^{(2)}$  and  $\chi^{(3)}$  are known as second and third order non-linear optical susceptibility.  $\chi^{(1)}$  is first order rank tensor,  $\chi^{(2)}$  is second rank and  $\chi^{(3)}$  is third rank tensor.

$$P^{(2)}(t) = \epsilon_0 \chi^{(2)} E^2(t)$$

$$P^{(3)}(t) = \epsilon_0 \chi^{(3)} E^3(t)$$

We also write that

$\chi^{(1)}$  is an order of unit

$$\chi^{(2)} \approx 1.94 \times 10^{-12} \text{ m/V}$$

$$\chi^{(3)} \approx 3.70 \times 10^{-24} \text{ m}^2/\text{V}^2$$

Consider an optical field incident upon a medium which is

$$E = E_0 \cos \omega t$$

Putting this value of  $E$  in eq<sup>n</sup> (1)

We get

$$P(t) = \epsilon_0 \left[ \chi^{(1)} E_0 \cos \omega t + \chi^{(2)} E_0^2 \cos^2 \omega t + \chi^{(3)} E_0^3 \cos^3 \omega t + \dots \right]$$

By using trigonometric relations

$$\cos 2\omega t = 1 - 2\cos^2 \omega t$$

$$\cos 3\omega t = -3\cos \omega t + 4\cos^3 \omega t$$

We get

$$P = \frac{1}{2} \epsilon_0 \chi^2 E_0^2 + \epsilon_0 \left( \chi + \frac{3\chi^3}{4} E_0^2 \right) \cos \omega t$$

$$\epsilon_0 \cos \omega t + \frac{1}{2} \epsilon_0 \chi^2 E_0^2 \cos 2\omega t$$

$$+ \frac{1}{4} \epsilon_0 \chi^3 E_0^3 \cos 3\omega t + \dots \quad \text{--- (2)}$$

The above equation clearly shows that presence of new frequency components due to non linear polarization.

In this equation first term is constant term due to DC field through the medium.

The second term is external polarisation also called as fundamental harmonic of polarization. The third term oscillates with frequency  $2\omega$  called as second

harmonic of polarization. The third term oscillates with frequency  $3\omega$  called as third harmonic of polarization and so on.

### Second order Term:

Second order process is proportional to product of two electric field.

$\omega = \omega_1 + \omega_2$  sum frequency mixing  
if  $\omega_1 = \omega_2$   $\omega = 2\omega$  Second Harmonic generation

if  $\omega_2 = 0$   $\omega = \omega$ , Pockel's effect

$\omega = \omega_1 - \omega_2$  difference frequency mixing

### Third order Term: It responses for

Third Harmonic generation

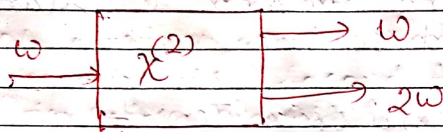
Raman effect

optical Kerr effect

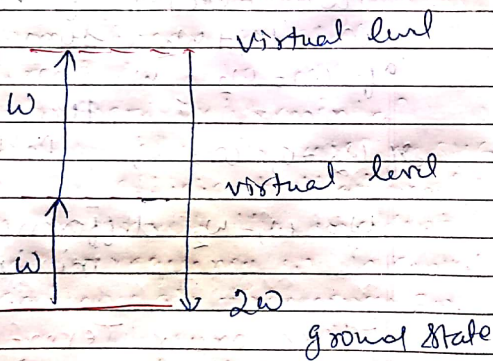
Four wave difference frequency mixing.



## Second Harmonic Generation (SHG)



### (a) Geometry of SHG



Energy level Diagram

Let us consider the process of Second Harmonic generation as an example of non-linear optical interaction shown schematically in above figure

In this case laser beam having electric field is represented by

$$E = E e^{-i\omega t} + \text{c.c.}$$

c.c. means Complex Conjugate

This electric field is incident upon a crystal for which second order susceptibility  $\chi^{(2)}$  is non zero.

The non linear polarization created in such crystal is given by

$$P^{(2)}(t) = \epsilon_0 \chi^{(2)} E^2(t).$$

OR

$$P^{(2)}(t) = 2\epsilon_0 \chi^{(2)} E E^* + (\epsilon_0 \chi^{(2)} E^2 e^{-2i\omega t} + \text{c.c.})$$

In this equation second order polarization consists of a contribution at zero frequency (First term) and a contribution at frequency  $2\omega$  (second term).



The second term contribution lead to generation of radiation of second harmonic frequency. The first term does not lead to generation of electromagnetic radiation because its second time derivative vanishes. This leads to a process called as optical rectification in which a static electric field created across the non-linear crystal.

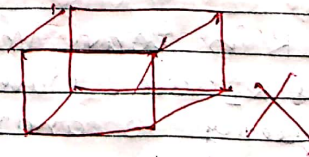
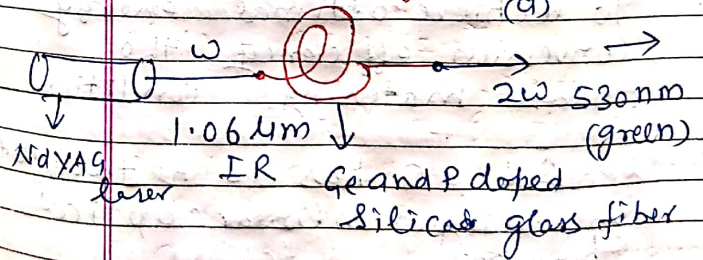
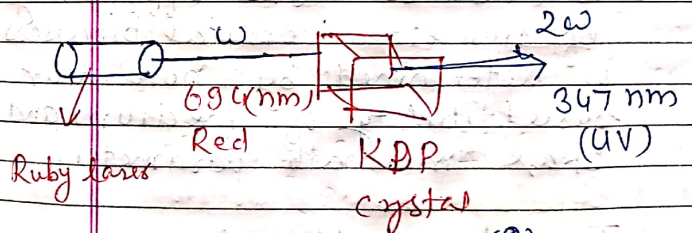
Under proper experimental conditions the process of SHG can be so efficient that nearly all the power in the incident beam at frequency  $\omega$  is converted to radiation at frequency  $2\omega$ .

From figure (a) two photons of frequency  $\omega$  are destroyed and a photon of frequency  $2\omega$  is simultaneously created. The solid line represents the atomic ground state while dashed lines represent virtual levels.

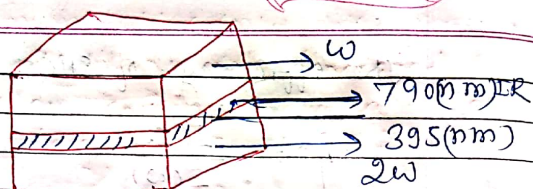
The efficiency of SHG

$$\eta = \frac{I(2\omega)}{I(\omega)}$$

Figures show several configuration for optical SHG in bulk materials and in waveguides in which Infra-red light is converted into visible and vice versa.







AlGaAs laser

**Phase matching:** In the process of SHG two photons having frequency  $\omega$  are destroyed and a photon of single frequency  $2\omega$  is created. The efficiency of non-linear optical processes is determined by nonlinear susceptibility of medium and phase mismatch parameter.

To achieve optimal energy conversion efficiency the phase matching condition

$\Delta K = 0$  has to be fulfilled in many non-linear optical processes.

Here  $K$  is wave vector.

If  $K_1$  and  $K_2$  are wave vectors for frequency  $\omega$  and  $2\omega$ .

then

$$K_1 = \frac{n_1 \omega}{c}, \quad K_2 = \frac{2n_2 \omega}{c}$$

$n_1 \rightarrow$  Refractive index

$n_2 \rightarrow$  " "

Momentum different

$$= \hbar |\Delta K|$$

$$= |\hbar K_2| - |\hbar 2K_1|$$

$$= \hbar (K_2 - 2K_1)$$

For Phase matching condition

$$K_2 - 2K_1 = 0$$

This is called Phase matching condition for second harmonic generation.

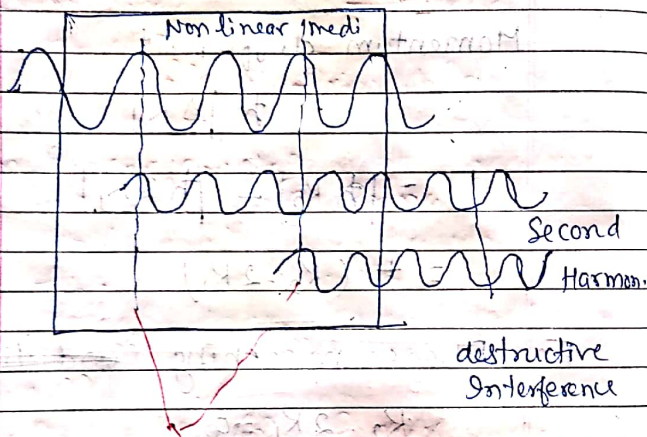
Consider the case of SHG

$$\omega_1 = \omega_2 = \omega$$

$$\omega_3 = 2\omega$$

A wave of frequency  $\omega$  propagates in the  $z$  direction through the crystal. At all  $z$  positions energy is transferred into a wave of frequency  $2\omega$ . For

maximum efficiency we require that all the newly generated light interfere constructively at the exit face of crystal.



Second Harmonic generation in phase to fundamental.

However dispersion within crystal means that fundamental and second harmonic light travel with different phase velocities. The phase matching condition is  $\Delta k = 0$  is equivalent to Conservation of momentum.

Phase matching are of following types

- (a) Phase matching by angle tuning
- (b) " " Temp. Tuning
- (c) Quasi Phase matching
- (d) Birefringent "
- (e) Walk off "
- (f) Noncritical "
- (g) Non collinear "

Optical Frequency Mixing

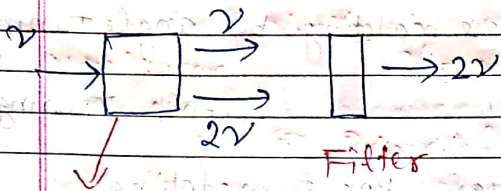
Optical Frequency Doubler:

A frequency doubler converts radiation of frequency  $\nu$  to radiation at doubled frequency. A filter blocks the radiation of frequency  $\nu$  that is not converted. The frequency doubler makes use of quadratic term of polarization

$$P = \epsilon_0 \chi^{(1)} E(\omega) + \epsilon_0 \chi^{(2)} E(\omega)$$

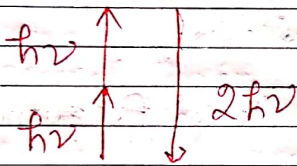


An electric field  
 $E = A \cos \omega t$



non linear crystal

(a) Principle



(b) Process

In an elementary process of frequency doubling, two photons of quantum energy  $h\nu$  are annihilated and a photon of energy  $2h\nu$  is created.

For examples

- (1) A frequency doubler converts infrared radiation to green radiation.

As nonlinear crystals  $\text{LiNbO}_3$  or KDP are suitable, the conversion efficiency can reach 40%.

- (2) Frequency doubling of green or blue radiation leads to UV radiation.

(a)

Difference Frequency generation

In a difference frequency generator, two sinusoidal fields of different frequencies  $\omega_1$  and  $\omega_2$  in which  $\omega_1 > \omega_2$  produce nonlinear polarization in a non linear crystal. The nonlinear polarization is source of an emf (electromagnetic field) at the difference frequency.

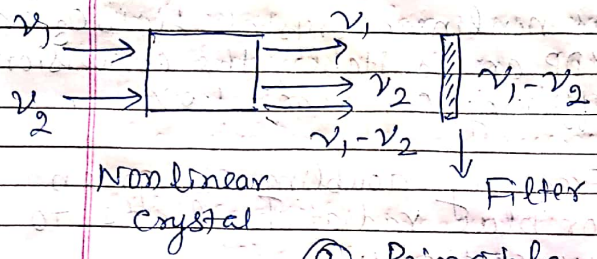
$$\omega_3 = \omega_1 - \omega_2$$

An electric field of frequency  $\omega_1$  and  $\omega_2$  is

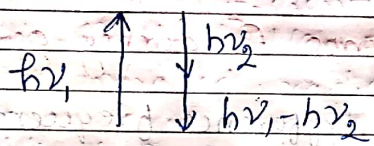
$$E_1 = A_1 \cos \omega_1 t$$

$$E_2 = A_2 \cos \omega_2 t$$





(a) Principle



(b) Process

Both electric fields superimposed to each other lead to the field

$$E = E_1 + E_2$$

This produces polarization

$$P = \epsilon_0 \chi^{(1)} (A_1 \cos \omega_1 t + A_2 \cos \omega_2 t) + \epsilon_0 \chi^{(2)} (A_1 \cos \omega_1 t + A_2 \cos \omega_2 t)^2$$

The polarization contains

the term

$$P_{\omega_1 - \omega_2} = \frac{1}{2} \epsilon_0 \chi^{(2)} A_1 A_2 \cos(\omega_1 - \omega_2) t$$

It means source of field is at different frequency. The field at the difference frequency  $\omega_1 - \omega_2$  are proportional to product of amplitude  $A_1$  and  $A_2$ . In an elementary process of difference frequency generation a photon ( $h\nu_2$ ) is annihilated and a photon ( $h\nu_3 = h\nu_1 - h\nu_2$ ) are created.

A photon of energy  $h\nu_1$  can only produce one photon of energy  $h\nu_3$ . This corresponds to energy conservation laws of elementary process. The efficiency of conversion of radiation at frequency  $\nu_1$  to radiation of frequency  $\nu_3$  is

$$\eta_{diff} = \frac{\nu_3}{\nu_1}$$

if  $\nu_3$  is much smaller



than  $\nu_1$  and  $\nu_2$ , only a small portion of power of radiation at frequency  $\nu_1$ , is converted to power of radiation at the difference frequency.

### Applications

The superposition of two visible or near infra-laser fields of different frequencies can lead to generation of far infrared radiation.

### Optical Parametric Oscillator

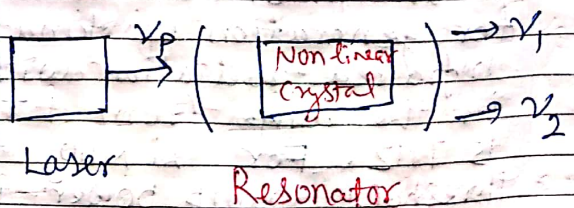
An optical parametric oscillator converts radiation of pump frequency ( $\nu_p$ ) to tunable radiation at two other frequencies ( $\nu_1$  and  $\nu_2$ ). Radiation of one of the frequencies  $\nu_1$ , or  $\nu_2$  or both is stored in a resonator in order to produce a feedback to the non linear crystal. The OPO shows threshold behaviour. Above a threshold amplitude of the pump

field, ~~OPO~~ set optical parameter oscillations set in. The oscillation frequency  $\nu_1$ , depends on the resonator. Changing the eigen frequency of the resonator lead to variation of  $\nu_1$  and  $\nu_2$ .

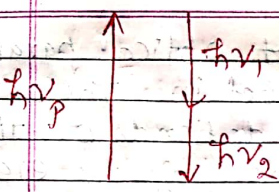
Phase matching can be achieved by the choice of appropriate orientation of non linear crystal for example  $\text{LiNbO}_3$ .

The elementary process in the optical parametric oscillator corresponds to decay of a photon into two photons of smaller quantum energy. The energy conservation law holds:

$$h\nu_p = h\nu_1 + h\nu_2$$





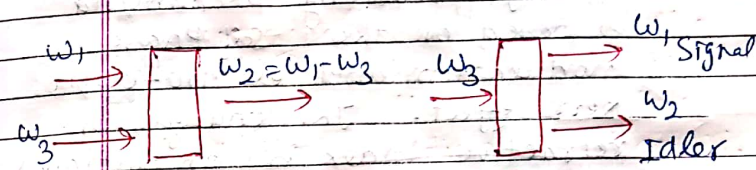


$\nu_p$  = Pump frequency  
 $\nu_s = \nu_i$  = Signal frequency  
 $\nu_2 = \nu_i$  = Idler Frequency

Parametric means that the pump field modulates a parameter and that parameter gives rise to frequency converting process. In the optical parameter oscillator is the refractive index of non linear medium

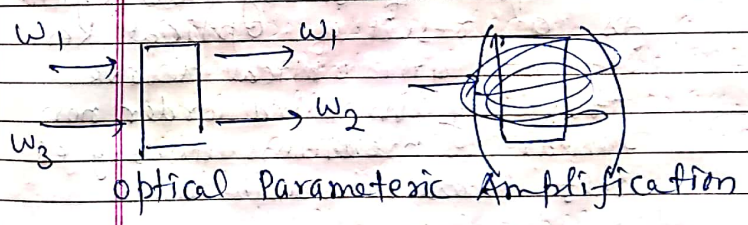
An OPO pumped with radiation near  $1 \mu m$  is suitable for generation of tunable Infra red radiation with frequencies in the range from 1 THZ to 15 THZ. For  $LiNbO_3$  the threshold pump power is order of  $1 MW/cm^2$ .

Diff. Frequency generation

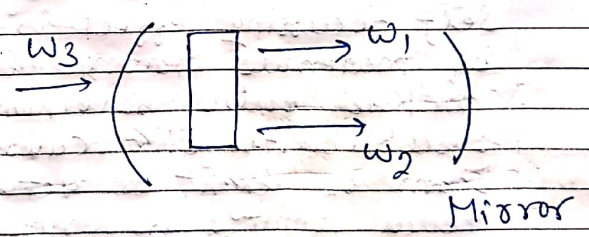


Diff. Frequency gene. Optical parameter generation (OPG)

Also called Parametric Down Conversion



Optical Parametric Amplification



Optical Parametric Oscillation



## Self Focussing Phenomena

Self focussing phenomena is based on the self phase modulation and electro-optic Kerr effect. The change in refractive index is proportional to the optical intensity. The overall refractive index is therefore a linear function of optical intensity. This is called Optical Kerr effect.

In electro-optical Kerr effect  $n$  (refractive index) is proportional to square of steady electric field. As a result of optical Kerr effect, an optical wave travelling in third order non linear medium undergoes self phase modulation.

Self focussing effect is exhibited by materials having large third order non linear optical susceptibility. This causes a beam of light to get focussed to a point in nonlinear optical medium without the

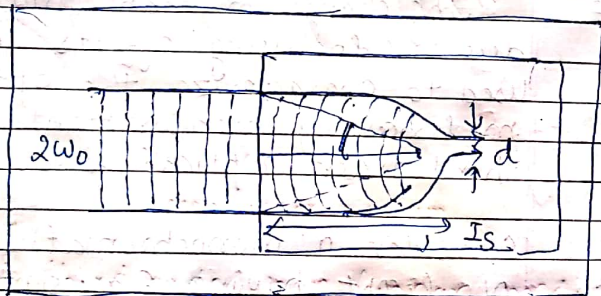
help of any external optical component like mirror or lens. Self focussing can be used to obtain an external narrow pencil of intense light.

The change in beam profile due to focussing effects can be used to characterize the optical material by measurement of third order optical susceptibility.

Consider a monochromatic beam of light having Gaussian cross section travelling through medium having negligible value of refractive index  $n_1$  (like air) and entering into transparent medium having significant value of refractive index  $n_2$ . The corresponding wavefront is plane parallel with air but starts getting distorted as beam begins to propagate in the medium. The intensity and refractive index are larger at centre of beam than at the periphery. Hence speed is



less at center. The plane wavefront bends shown in figure, leading to focussing in a medium with  $n_2 > 0$ .



angle of focussing =  $\theta_s$

The parameters that describe self focussing are

- (i)  $w_0$  the Beam radius before it enters in non linear medium
- (ii)  $\theta_s$  the angle of self focussing
- (iii)  $d$  minimum Beam diameter on self focussing within a medium of refractive index  $n_0$
- (iv) The non linear refractive index  $n_2 I$ ,  $I$  is intensity of beam

The angle of self focussing

$$\theta_s = \sqrt{\frac{2n_2 I}{n_0}}$$

and distance  $I_s$  at which beam focusses is given by

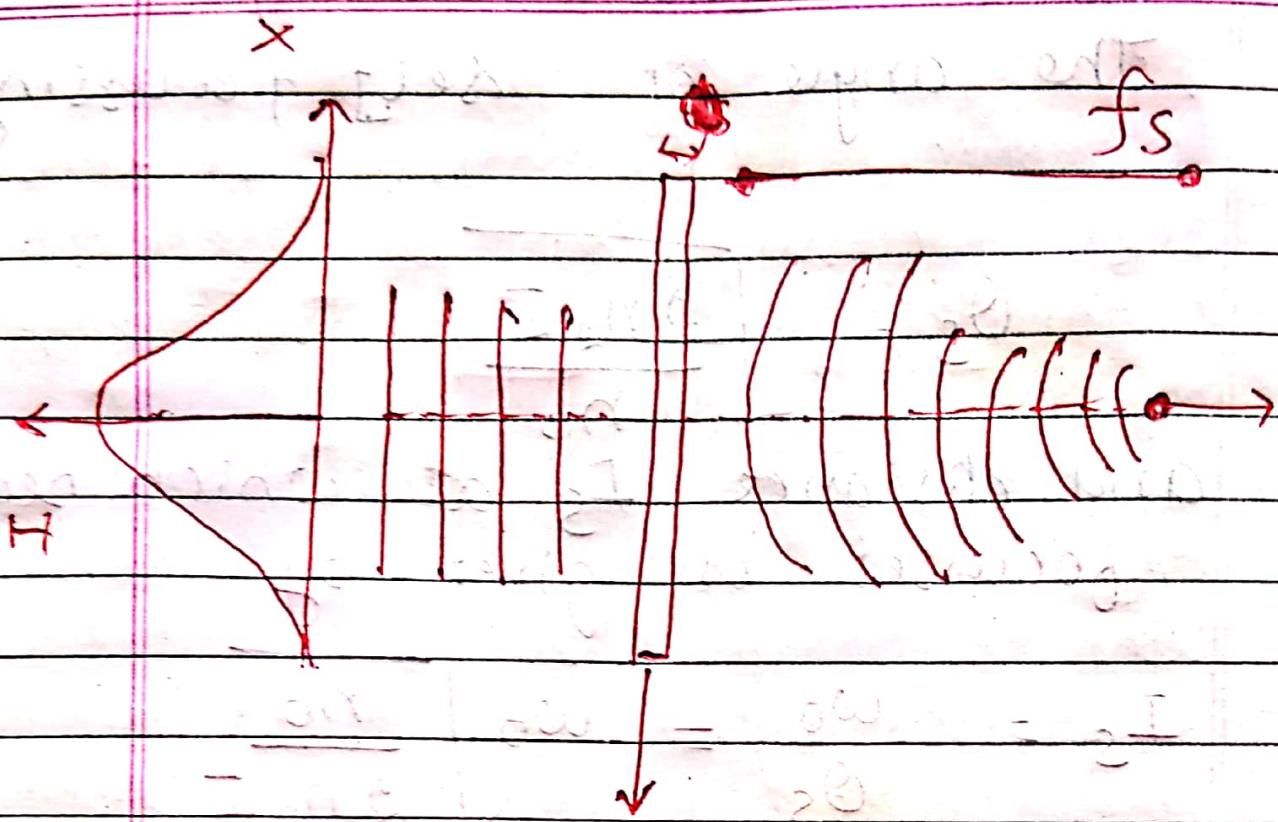
$$I_s = \frac{w_0}{\theta_s} = w_0 \sqrt{\frac{n_0}{2n_2 I}}$$

From above equation,  $\theta_s$  is ~~not~~ directly proportional to square root of nonlinear refractive index. The corresponding focal length  $I_s$  is inversely proportional to it.

It may be noted that some materials have negative value for  $n_2$  which leads to self defocussing effect and divergence of beam instead of self focussing.

The self focussing can also be shown in figure





non linear medium

A third order non-linear medium acts as a lens having focal length  $f_s$  which depends upon intensity of incident beam