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Experimental Study of Third Order Nonlinear Absorption in Pure and MG Doped Lithium Niobate Crystals

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ABSTRACT

With the discovery of laser in 1960 various nonlinear effect arises, the origin of nonlinear optics lies in the nonlinear response of materials to the incident coherent radiation. Using pulsed Q-switched Nd: YAG Laser, we have observed the important phenomena i.e. third-order nonlinear effect in the LiNbO3 crystals. In present work, I have performed an experiment using simple and sensitive single beam Z-scan technique to measure nonlinear absorption in LiNbO3 crystals samples such as pure LiNbO3 crystal, 5mol% Mg doped LiNbO3 crystal, and 7mol% Mg doped LiNbO3 crystal.

KEYWORDS: *Nonlinear coefficient, Optical phase conjunction, Nonlinearity, Third harmonic generation*

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INTRODUCTION TO NLO

The Physical phenomena related to light and its propagation is broadly classified as "Optics". Basically optics described the generation, composition, transmission and interaction of light. This is primarily due to advent of LASER as a primary source of power in 1960 [1].The field originated with the experimental work of P.A. Franken and coworkers[2] on "Optical SHG" in 1961 and theoretical work of N. Bloembergen and coworkers in 1962 [3]. In pre-Laser era people thought that optical properties of medium are independent of light incident intensity of radiation. The reason behind this conclusion is that the strength of atomic and interatomic field (10^7 to 10^{10} v/cm) and such low field cannot influence has the field strength in the range of 10^8 v/cm, affects the inter atomic effect which correspond gives the nonlinear effects. OPC offers the possibility of all signals processing, with the advantages of greater speed, integration with factor technology and possibility of parallel computing of this [4]. The third-order susceptibility is responsible for the process like Third Harmonic Generation (THG), Self-focusing, Optical Phase Conjugation, Two photon absorption, Optical Kerr effect, Self Phase Modulation and Intensity dependent absorption and refraction and so on. Symmetric materials do not exhibit even order nonlinearities [5]. To this end, a simple and sensitive single beam technique referred to as "Z-scan" has been described for the measuring the nonlinear absorption for a wide variety of materials. Other techniques which have been used for the nonlinear absorption in materials are: degenerate four-wave mixing, nearly degenerate three-wave mixing, and Kerr effect with two photon absorption. The original technique given by Sheik-Bahae has been extended to various materials for the measurement of nonlinear absorption co-efficient [6]. The Z-scan technique provides simplicity as well as high senility for measuring nonlinear absorption and refraction in solids and liquid samples. This single beam Z-scan method is based on the self-focusing and self-defocusing phenomena in optical nonlinear materials. In the Laser beam is focused by a lens to at Z=0 point along the propagation direction of beam (Z-direction) as shown in figure (2.1). Basically sample is translating the through the focal plane of tightly focused laser beam and monitoring the changes in the far field intensity pattern [7].

ORIGIN OF OPTICAL NONLINEARITY:

When the applied electric fields are small, the electric polarization **P** is approximately linearly proportional with the applied electric field **E**

$\mathbf{P} = \epsilon_0 \chi^{(1)} \mathbf{E}$ in SI.

$= \chi^{(1)} \mathbf{E}$ in cgs. (1)

Relation (1) shows that **P** is linearly proportional to **E**. ϵ_0 being the electric permittivity of free space and since the electric field is associated with a light beam, when a light beam propagates through a dielectric medium it also induces

electric polarization as shown in equation (1) and hence **P** is linearly proportional to **E.** Polarization **P** of the medium can be expressed as a power series in **E**.

$\mathbf{P} = \epsilon_0 \chi_{eff} \mathbf{E}$ (2)

$\mathbf{P} = \epsilon_0 (\chi^{(1)} + \chi^{(2)}\mathbf{E} + \chi^{(3)}\mathbf{E}\,\mathbf{E} \ldots\ldots\ldots\ldots\ldots) \mathbf{E}$ (3)

Where χ is nonlinear with respect to the field strength of the light wave.

Various The induced polarizations as given by Eq. (3) can be expressed as

$\mathbf{P} = \mathbf{P}^{(1)} + \mathbf{P}^{(2)} + \mathbf{P}^{(3)} + \ldots\ldots\ldots\ldots\ldots\ldots$ (4)

Here, $\mathbf{P}^{(1)}$ is the polarization due to fist power of **E**, $\mathbf{P}^{(2)}$ is polarization due to $\mathbf{E}^2$ term hence is called quadratic polarization, $\mathbf{P}^{(3)}$ is due to $\mathbf{E}^3$ and is called cubic polarization more popularly, they are known as second , third-order polarization. $\chi^{(1)}$, $\chi^{(2)}$ and $\chi^{(3}$are the optical susceptibilities. Any medium whose polarization is given by above form is called as a nonlinear polarization. For a medium with inversion symmetry the polarization is given by

$\mathbf{P} = \epsilon_0 (\chi^{(1)}\mathbf{E} + \chi^{(3)}\mathbf{EEE} \ldots\ldots\ldots\ldots\ldots)$ (5)

Such medium will have, in addition to linear term, only "third-order nonlinearities" as well as higher order nonlinearity while for noncentrosymmetric (NCS) system the cubic term is substantially smaller than $\mathbf{P}^{(2)}$. The total polarization for such system is expressed as

$\mathbf{P} = \mathbf{P}^{(1)}\mathbf{E} + \mathbf{P}^{(2)}\mathbf{E}^2 \ldots\ldots\ldots\ldots\ldots\ldots$ (6)

Such a medium is said to have "second-order optical nonlinearities".

Third Order Nonlinearity

Let us consider a medium in which the optical nonlinearities arise due to the third and higher odd-order term, and then the induced polarization at the field frequency ω has the form.

$P(\omega) = \frac{1}{2} [P^L(\omega) + P^{NL}(\omega)]$ +Complex conjugate (C.C.) (7)

The superscript L stands for the linear while NL for the nonlinear component. For a monochromatic light wave, E can be represented as

$E = \frac{1}{2} [E_0 e^{-i\omega t} + E_0^* e^{i\omega t}]$ (8)

Using (7) and (8) restricted only to the third-order nonlinear terms, the polarization at frequency ω takes form

$P(\omega) = \frac{1}{2} \epsilon_0 \chi_{eff} E_0 e^{-i\omega t}$ +C.C.+…….. (9)

Here, we define the effective optical susceptibility of the medium χ_{eff} as

$\chi_{eff} = \chi^{(1)} + \chi^{(3)} | E_0(\omega)|^2$ (10)

Where $|E(\omega)|^2 = E_0(\omega) E_0^*(\omega)$

In general, the co-efficient of proportionality between P(ω) and E(ω) determines the refractive index n of the medium given by

$n(I) = n_0 + n_1 I$ (11)

Where, the linear refractive index is expressed as

$n_0^2 = 1 + \chi^{(1)}$ (12)

Therefore third order nonlinear susceptibility is now considered to be a complex quantity:

$\chi^{(3)} = \chi_R^{(3)} + i \chi_I^{(3)}$ (13)

Where imaginary part is related to two photon absorption coefficient β though

$\chi_I^{(3)} = n_0 \epsilon_0 c^2 / \omega \beta$

And real part is related to nonlinear refraction coefficient γ though

$\chi_R^{(3)} = 2 n_0^2 \epsilon_0 c \gamma$ (14)

Therefore we can define nonlinear coefficient in following way

$\alpha(I) = \alpha_0 + \alpha_1 I$ (15)

Where α_0 is the linear absorption coefficient and α_1 is the nonlinear absorption coefficient and (I) is the Intensity of Laser beam.

DESCRIPTION OF THIS TECHNIQUE:

The Z-scan experimental arrangement is given below

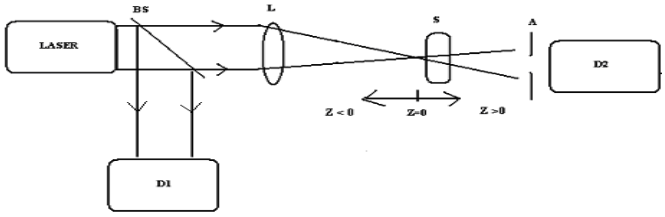


Figure 2.1: Experimental arrangement of single beam Z-scan technique.

LS = Laser Source. , **BS =** Beam splitter, **S** = Sample

L = Focusing Lens**. A =** Aperture. **D_1 =** Reference detector**.**

D2 = Second detector**. Z =0** Shows the distance at focus. **Z > 0** Shows positive Z direction towards detector.**, Z < 0** Shows negative z direction towards lens**.**

The Z-scan technique is a sensitive and simple experimental technique to measure intensity dependent optical nonlinear susceptibility like nonlinear absorption and nonlinear refraction of material. In this technique, the sample is translated in Z-direction along the axis of focused Gaussian beam, and the far field intensity is measured by detector placed in far field. The far field intensity is measured as function of sample position Z, shown in the figure (2.1). After the focal plane, the same self-defocusing increases the beam divergence, leading to a widening of the beam at the iris and thus reducing the measured transmittance. Far from focus ($z > 0$), again the nonlinear refraction is low resulting in a transmittance *z*-independent. A pre-focal transmittance maximum (peak), followed by a post-focal transmittance minimum (valley) is a Z-scan signature of a *negative nonlinearity*. Inverse Z-scan curves (i.e., a valley followed by a peak) characterize a *positive* nonlinearity, as shown in the figure (2.2)

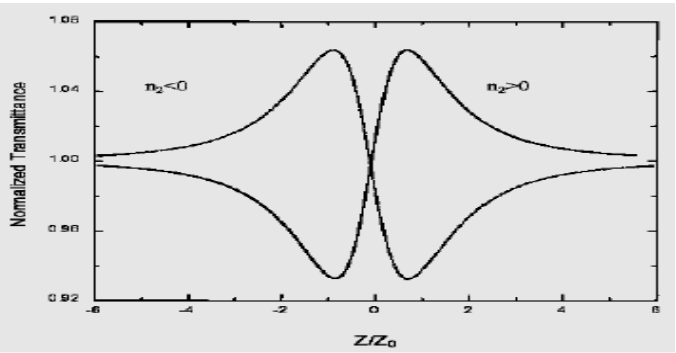


Figure 2.2: Z-scan characteristics for thin self-defocusing and self-focusing medium.

Now we discuss about the mechanism for study of nonlinear absorption is called the open aperture Z-scan technique. Let us consider a thin sample having nonlinear absorption coefficient (α). After remove the aperture then all the transmitted light come on the detector, this results in a flat response for pure refractive nonlinearity. If nonlinear absorption is present, then the transmittance signal has a minimum at Z=0 (The sample at the focal plane) where the irradiance is maximum as shown in figure (2.3). Nonlinear absorption suppresses the peak and enhances the valley in Z-scan.

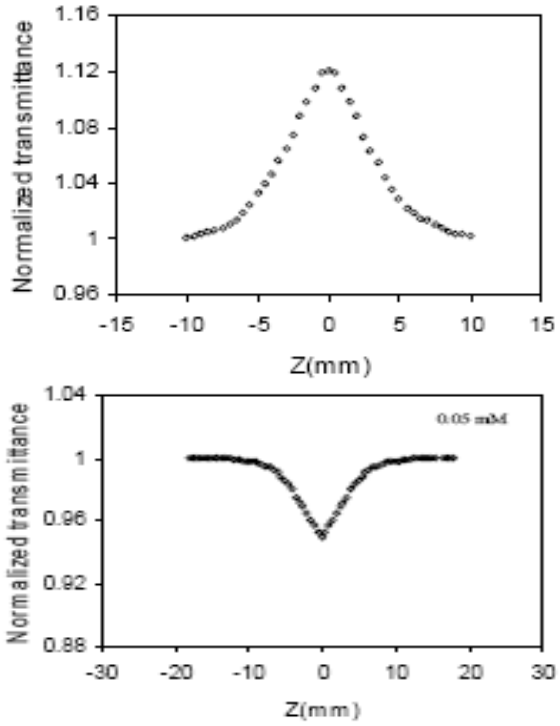


Figure 2.3: Z-scan Characteristics (saturation absorption and two photon absorption) for thin absorber medium.

EXPERIMENTAL SET UP:

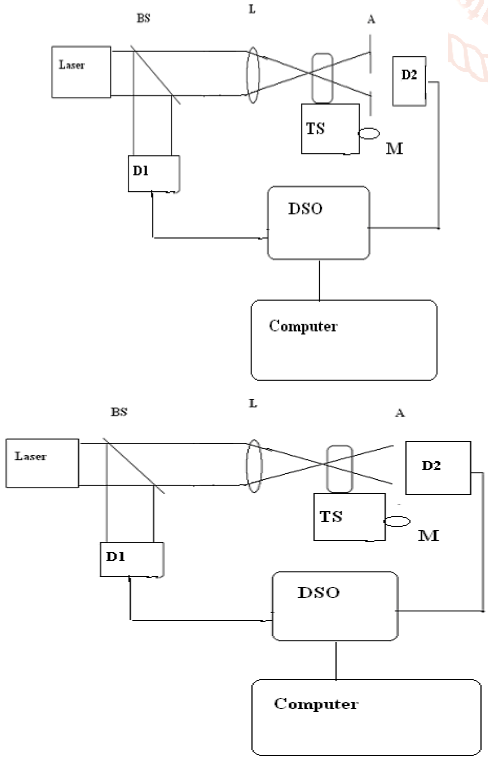


Figure3.1, 3.2: Experimental set up of closed & open aperture Z-Scan Technique

Laser = Q Switched Nd: YAG 1.064 μm Laser.

BS = Beam Splitter, **S** = Sample, **L** = Focusing Lens. **M** = Stepper motor **A** = Aperture **D1 & D2** = Photo detector.

DSO = Digital Storage Oscilloscope. **TS** = Motorized Translation Stages, **Computer** = Shows the computer with Lab View software

We have adopted the simple and well known M. Sheik-Bahae open aperture Z-scan setup for the measurement of nonlinear absorption coefficient α_1. The experimental setup of open aperture Z-scan technique is shown in figure (3.2).

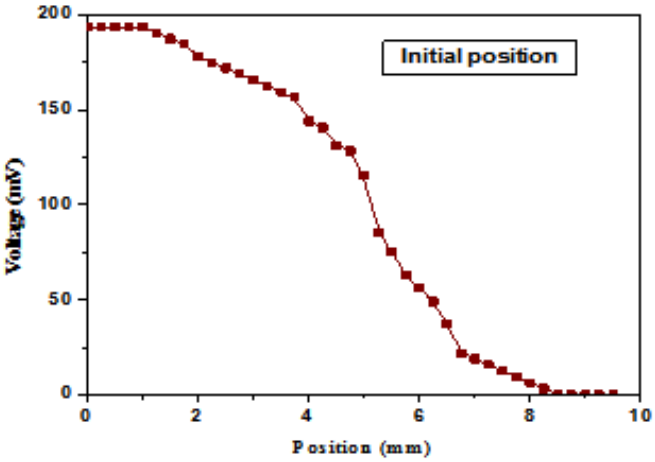
EXPERIMENTAL OBSERVATION

In the present work, we have carried out investigation of nonlinear absorption coefficient α_1 in $LiNbO_3$ crystals samples (such as pure $LiNbO_3$ crystal, 5% mol Mg doped $LiNbO_3$ crystal and 7%mol Mg doped $LiNbO_3$ crystal) by using the open aperture Z-scan technique, at the fundamental wavelength 1.064μm Nd: YAG laser. The following task have been carried out,

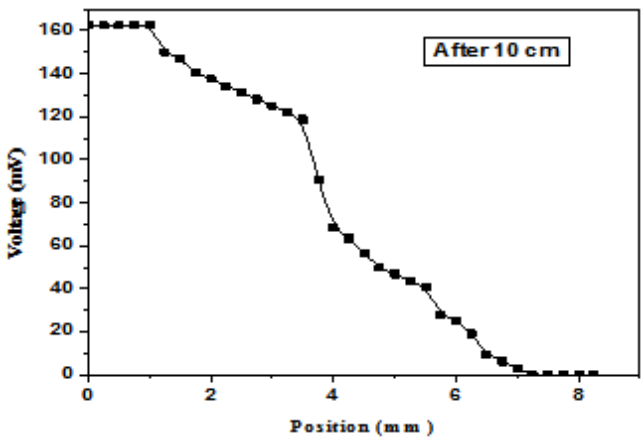
1. **Measurement of spot size:**

For the measurement of spot size, we have used knife-edge, focusing lens, DSO and detector. The experimental set up for spot size measurement is shown in figure (4.1). \

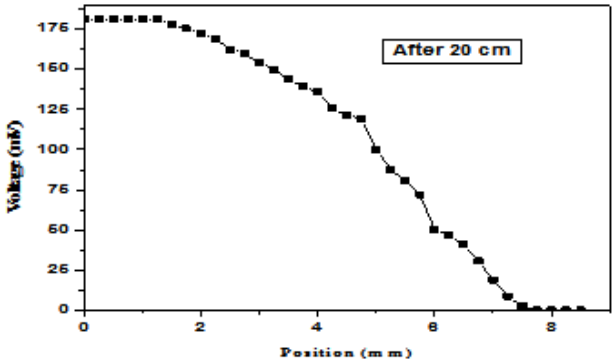
A. **At initial position from laser source** Thus spot size corresponding initial position (10 cm from laser source) will be Spot Size = 6.643 mm



Graph 4.1: Curve for initial position.



Graph 4.2: Curve for after 10 cm from initial position.



Graph 4.3: Curve for 20 cm from initial position.

B. After 10 cm from the initial position: The spot size for after 10 cm from initial position will be Spot size = 6.751 mm shown in graph 4.2

C. After 20 cm from the initial position: spot size corresponding to after 20 cm from initial position Spot size = 6.787mm shown in graph 4.3

2. Measurement of beam waist W_0 (spot size at the focus):

$\theta_0 = 1/\sqrt{2D}\,(W_3^2 - 2W_2^2 + W_1^2)^{1/2}$ (15)

Thus θ_0 is measured in term of experimentally beam radii, and we also measured

$W_0 = \lambda / \pi\,\theta_0$ (16)

From above equations we calculate W_0

Calculation for beam waist W_0

We have W_1 = 6.843 mm W2 = 6.751m W_3 = 6.787mm

After using equation (15) we obtain

$\theta_0 = 0.0093$

Now from equation (16), we obtain beam waist is

Beam waist W_0 = 46 µm (spot size at the focus)

3. Measurement of linear absorption coefficient:

Linear absorption coefficient is obtained by following equation

$\alpha_0 = -1/L \log (V/V_0)$ (17)

Where V= Voltage with sample. V_0 = Voltage without any sample.

The values of linear absorption coefficient for three $LiNbO_3$ samples are given in the below table (4.1)

S. No.	Sample	Wavelength (in µm)	Voltage (V_0),voltage without sample (mV)	Voltage(v) with the sample (mV)	α_0 in cm^{-1}
1	Pure $LiNbO_3$	1.064	475	450	0.54
2	5 mol % doped $LiNbO_3$	1.064	475	443	0.69
3	7 mol % doped $LiNbO_3$	1.064	475	437.5	0.82

Table 4.1: Values of linear absorption coefficient for three $LiNbO_3$ samples

4. Measurement of effective length of samples:

The effective length of the sample is given by,

$L_{eff} = 1\text{-}\exp(-\alpha_0 L)/(-\alpha_0)$ (18)

Where α_0 = linear absorption coefficient of the sample. L = true sample length. The values of effective length of our three LiNbO3 samples are given in the below table (4.2)

S. No	Sample	True sample length (cm)	Linear absorption coefficient α_0 (cm^{-1})	Effective length (in cm)
1	Pure $LiNbO_3$	0.1	0.54	0.0973
2	5 mol % $LiNbO_3$	0.1	0.69	0.0966
3	7 mol % $LiNbO_3$	0.1	0.82	0.0960

Table 4.2: Values of effective length of our three LiNbO3 samples

5. Measurement of nonlinear absorption coefficient:

For measuring nonlinear absorption coefficient, we have used curve fitting analysis. The Normalized Transmittance in the case of open aperture condition is given by

$T(z) = \ln(1+q_0)/q_0$ (19)

Where q_0 is given by following equation,

$q_0 = \alpha_1 . I_0 [1-\exp(-\alpha_0 L)] / [1 + (z/z_0)^2].\alpha$ (20)

Here α_0 = Linear absorption coefficient of the sample.

L = Sample Thickness, I_0 = Intensity of laser beam at the focus,

α_1= is the nonlinear absorption coefficient of the sample.

Let us assume that the efficient length approximately equal to length of our sample, in this condition equation (3.10) is given by

$q_0 = \alpha_1 . I_0 L / [1+ (z/z_0)^2]$ (21)

$$T(z) = \frac{\ln(1+q_o)}{q_o} = \frac{(1+(\frac{z}{z_0})^2)\ln(1+\frac{2}{(1+(\frac{z}{z_0})^2)})}{2}$$

Now after knowing value of q_0, I_0, L we can determine value of nonlinear absorption coefficient α_1 by following equation,

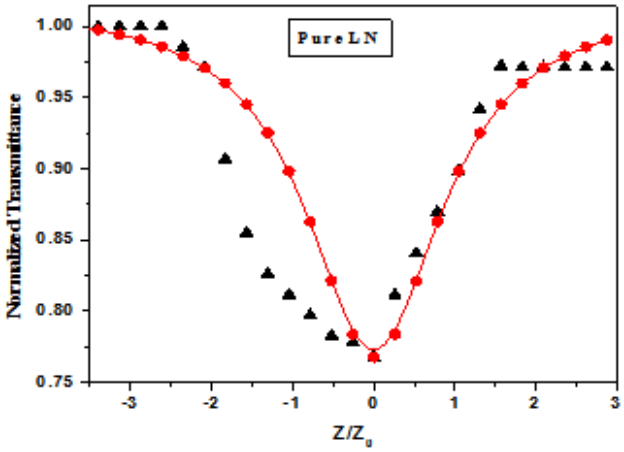
$\alpha_1 = q_0 / (I - L_{eff})$ (22)

Where L_{eff} = effective sample length.

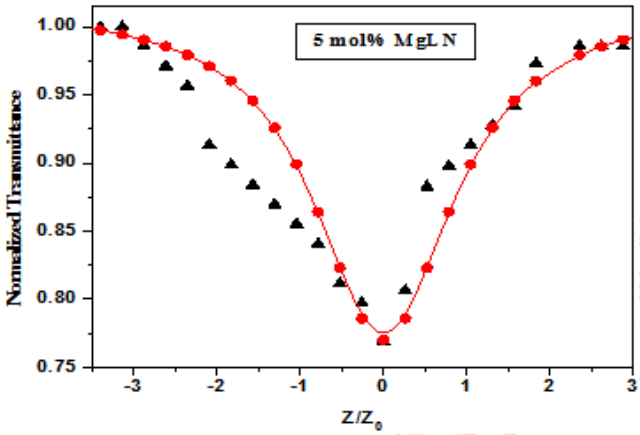
6. Calculation for nonlinear absorption coefficient (α1):

I. For pure $LiNbO_3$ crystal:

In the graph line curve shows the theoretical value (fit) and scattered point curve shows experimental value (fit).



Graph 4.4: Curve for Pure $LiNbO_3$ crystal.



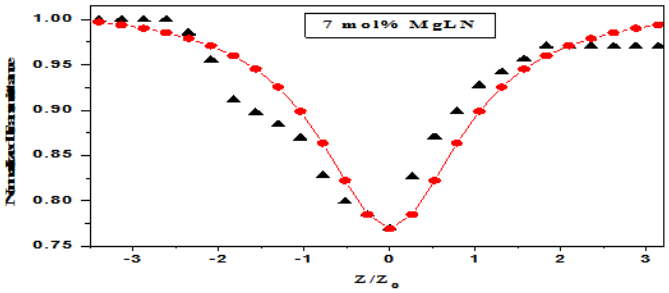
Graph 4.5: Curve for 5mol % Mg doped $LiNbO_3$ crystal.

We have L = 0.1 cm (sample thickness).
α_0 = 0.54 cm^{-1} (linear absorption coefficient).
I = 0.26 x 10^9 (Intensity at the focus)
L_{eff} = 0.09735 cm. (effective sample length).
q_0 = 0.6935. $\alpha_1 = q_0 / (I - L_{eff})$
Thus from above equation we obtain, α_1 = 2.74 x 10^{-8}

II. For 5 mol % Mg doped LiNbO3 crystal:
For 5mol % Mg doped $LiNbO_3$ crystal and we obtained fitted curve between Normalized Transmittance and Sample Thickness is shown in the graph (4.5).

We have L = 0.1 cm (sample thickness).
α_0 = 0.69 cm^{-1} (linear absorption coefficient).
I = 0.26 x 10^9 (intensity at the focus)
L_{eff} = 0.09663 cm. (effective sample length).
q_0 = 0.6868 $\alpha_1 = q_0 / (I - L_{eff})$
From above equation we obtain, α_1 = 2.734 x 10^{-8}

III. For 7 mol% Mg doped LiNbO3 crystal:
For 7mol% Mg doped $LiNbO_3$ crystal, we obtained fitted curve between Normalized Transmittance and Sample Thickness is shown in the graph (4.6).



Graph 4.6: Curve for 7mol % Mg doped $LiNbO_3$ crystal

We have L = 0.1 cm (sample thickness).
α_0= 0.82 cm^{-1} (linear absorption coefficient).
I = 0.26 x 10^9 (intensity at the focus)
L_{eff} = 0.09601 cm (effective sample length).
q_0 = 0.6868. $\alpha_1 = q_0 / (I - L_{eff})$
From above equation after putting all values, we obtain α_1 = 2.764 x 10^{-8}

EXPERIMENTAL RESULTS:

A. Results for Pure $LiNbO_3$ crystal:
While studying pure $LiNbO_3$ crystal sample, we found that linear absorption coefficient α_0, which determined experimentally is = 0.54 cm^{-1} and effective length for pure $LiNbO_3$ crystal sample is = 0.09735 cm. From curve fitting we found that value nonlinear absorption coefficient α_1 = 2.74 x 10^{-8}. Thus a theoretical curve is corresponds to experimental curve. The curve for pure $LiNbO_3$ crystal is shown in the graph (4.4).

B. Results for 5mol % Mg doped LiNbO3 crystal:
After studying 5mol% $LiNbO_3$ crystal sample, we have found that linear absorption coefficient α_0 = 0.69 cm^{-1} and we have also determined effective length for 5 mol% $LiNbO_3$ crystal sample is = 0.09663 cm. The value of nonlinear absorption coefficient α_1 is = 2.734 x 10^{-8}. Thus a theoretical curve is corresponds to experimental curve. The curve for 5 % mol $LiNbO_3$ crystal shown in the graph (4.5).

C. Results for 7 mol % Mg doped LiNbO3 crystal
After studying 7mol % Mg doped $LiNbO_3$ crystal sample, we found that linear absorption coefficient α_0 is = 0.82 cm^{-1}, effective length for sample is = 0.09601cm. For measurement of nonlinear coefficient, we have used curve fitting analysis from curve fitting analysis we found that value nonlinear absorption coefficient is α_1 = 2.764 x 10^{-8}. Thus a theoretical curve is corresponds to experimental curve. The curve for 5mol % $LiNbO_3$ crystal shown in the graph (4.6).

CONCLUSION:
Nonlinear absorption coefficient in Lithium niobate crystal ($LiNbO_3$) depends on Mg doping concentration and optical threshold value of the crystals. In pure Lithium niobate crystal we obtained higher values of nonlinear absorption coefficient and for 7mol % $LiNbO_3$ crystal we obtained again higher value similar to pure $LiNbO_3$ crystal. And for 5mol % $LiNbO_3$ crystal we obtained smaller nonlinear absorption coefficient value as compared to Pure LiNbO3 crystal and 7 mol% LiNbO3 crystals. All our experimental value corresponds to reported values. It is found that nonlinear absorption coefficient also depends on the Mg doing concentration in the crystal. If doping concentration is large, and then we obtain smaller values of nonlinear absorption because of larger light observed by crystal, and experimental curve corresponds to theoretical curve.

Future Scope:
In the present work we have studied the nonlinear absorption coefficient in pure $LiNbO_3$ crystal, by curve fitting analysis and we also study the linear absorption coefficient, effective sample length. And we have studied spot size at three positions from laser. We also determined beam waist W_0 for laser beam. Similar technique can be used for measurement of nonlinear refractive index; it is useful for nonlinear devices application. The single beam Z-Scan technique is attractive owing to its experimental simplicity

and sensitivity and that it yields both sign and the magnitude of nonlinearity. A further advantage is close similarity Z-Scan and optical power limiter geometries. the Z-Scan technique not only gives important information on nonlinear optical characteristics of material but also yield vital information regarding optimization of optical power limiter geometry such as optimum sample thickness and optimum sample position. The Z-scan technique can also be used with different laser polarization conditions, to obtained information about the different tensor component of χ^3 .It can be employed for studying third order nonlinear optical effect in different organic molecular materials. Lithium niobate is used extensively in the telecoms market, e.g. in the mobile telephones and optical modulators.

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