Study of the Nonlinear Optical Properties of Lithium Triborate Crystal by Using Z-Scan Technique

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Abstract. ***In this experimental work, a study (by using a high sensitive method known as Z-Scan technique) of the nonlinear optical properties for Lithium Triborate crystal (The boron–oxygen compound LiB_3O_5 is recognized as a new Nonlinear Optical crystal).Z-scan experiment was performed using Nd-Yag laser at 532 and 1064 nm with the power output of 40 and 25 mW, in two parts. The first part was done using a closed-aperture placed in front of the detector to measure the nonlinear refractive index. In the second part; the aperture in front the detector was removed (open aperture) to measure the nonlinear absorption coefficient. The experimental results for closed aperture show that the Lithium Triborate crystal exhibits positive refractive index (self-focusing). For open aperture z-scan the results show that the crystal exhibited saturation absorption.***

Keywords: Z-scan technique, nonlinear refraction and nonlinear absorption coefficient, Lithium Triborate

1. Introduction

Nonlinear optics (NLO) is the study of all the phenomena that occur from the interaction of intense light with matter [1, 2]. The interaction with a nonlinear optical material causes a modification of the optical properties of the system, and the next photon that arrives sees a different material. Typically only laser light is sufficiently intense to generate NLO phenomena; therefore the beginning of this research field is often taken to be the discovery of second-harmonic generation by Franken *et al.* in 1961[3], the year after the construction of the first laser by Maiman [4].Although the observation of most nonlinear optical phenomena requires laser radiation, some classes of nonlinear-optical effects were known long before the invention of the laser. The most prominent examples of such phenomena include Pockel's and Kerr electro optic effects[5].The theory of nonlinear optics builds on the well-understood theory of linear optics, particularly that part known as the interaction of light and matter. Ordinary matter consists of a collection of positively charged cores (of atoms or molecules) and surrounding negatively charged electrons Light interacts primarily with matter via the valence electrons in the outer shells of electron orbitals. The fundamental parameter in this light-matter interaction theory is the electronic polarization of the material induced by light. [6]

2. Literature Review

2.1 Characteristics of Lithium Triborate crystal

The existence of Lithium Triborate (LiB_3O_5) was already found in 1926 by Mazetti and Carli. However, it took until 1989 that the excellent nonlinear properties of LBO crystals were discovered [7].The LBO crystal is a negative biaxial crystal, it belongs to the point group mm2. The crystal structure is based on a large number of anionic boron-oxygen bonds $(B_3O_7)^{5-}$ organized around Lithium (Li^+) cations. It is the anionic group, which is assumed to be responsible for the basic nonlinear properties of Borate crystals as LBO, Because of LBO crystal's high optical damage threshold (for damage in the crystal bulk) and being highly transparent from the near-infrared to the near ultraviolet [8].

This crystal considers to be one of the most important nonlinear optical crystals for frequency conversion because of its excellent optical properties, such as [9]:

1) The second-order nonlinear optical coefficient is large (d_{eff} 50.96 pm/V).
2) The laser damage threshold is higher about (25 GW/cm^2, 1.064 mm, 0.1 ns) makes LBO crystals very suitable for harmonic generations of high-intensity laser radiation in a wide spectrum.
3) Broad transparency range (0.16–3.5 mm), and good chemical and mechanical stability.
4) Its physical properties, such as infrared reflectance, Raman spectra, elastic properties, and dielectric properties, as well as its optical properties.[9] (as shown in table 1).

Table 1: Crystal Structural, Physical and chemical Properties [10]

Chemical Formula	LiB_3O_5
Crystal Structure	Orthorhombic, Space group Pna2$_1$, Point group mm2
Lattice Parameter	a=8.4773Å, b=7.3788Å, c=5.1395Å, Z=2
Melting Point	About 834°C
Mohs Hardness	6
Density	2.47 gm/cm^3
Absorption coefficient	< 0.1%/cm (at 1064nm and 532nm)
Specific heat	1.91J/cm^3.K, 1060 J/kg.K
Hygroscopic susceptibility	Low
Damage threshold	25 J/cm^2 (1064 nm, 10 ns pulses)
Thermal Conductivity	3.5W/m/K
Thermal Expansion Coefficient	α_x=10.8x10^{-5}/K, α_y= -8.8x10^{-5}/K, α_z=3.4x10^{-5}/K

2.2 The Z-Scan Technique

In 1990 M. Sheik-Baha[11] established a measurement method with the purpose of determining the nonlinear refraction of thin samples. This is called the Z-scan technique[12]. It is a single-beam technique that gives us both the sign and magnitude of refractive index nonlinearities and Nonlinear absorption, which are associated with the real part $\chi_R^{(3)}$ and imaginary part $\chi_I^{(3)}$ of the third order Nonlinear susceptibilities[13]. The Z-Scan technique has been used to measure the Nonlinear optical properties of semiconductors, dielectrics, organic or carbon-based molecules and liquid crystals. [14] This method is rapid, simple to perform and accurate, therefore it is often used. It is especially adequate for determination of a nonlinear coefficient n_2 for a particular wavelength. The essential geometry is shown in Fig.1[15]

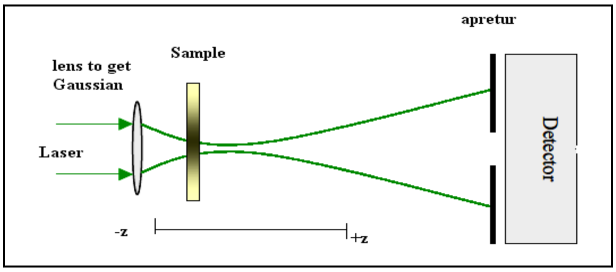

Figure 1: Z-scan experimental arrangement

The Z-Scan technique include that the sample is scanned along the Z direction through the beam waist of a focused Gaussian laser beam in a tight focusing configuration, as seen in Figure 2. As the sample approaches focus the spot size decreases, increasing the irradiance on the sample and the induced nonlinear effects.[16]

There were two parts of the Z-scan: Closed Aperture and Open Aperture.

2.2.1 Closed aperture Z- Scan

Closed aperture Z-scan is an example of self-refraction phenomenon or self-phase modulation in space. In the absence of nonlinear absorption, a well-defined peak and valley are observed. If the nonlinear refractive index n_2 of the sample is negative, the beam gets converged in the pre-focal region to get focused closer to the aperture. Consequently, the beam diameter decreases near the aperture, resulting in large amount of through put at the detector.[17] This results in a peak in the pre focal region. In the post focal region, the same phenomenon results in the divergence of the beam, which results in the decreased transmission through the aperture. Hence, a valley appears in the post-focal region. If the sample has positive nonlinear refraction, we have just the opposite result (pre focal valley and post focal peak.). The former is called self-defocusing and the latter is called self-focusing. One of the mechanisms of self-focusing is optical Kerr effect, which has instantaneous response. In this case the electric field of a light beam exerts a torque on anisotropic molecules by coupling to oscillating dipole induced in the molecule by the field itself. Resulting light induced molecular reorientation is the main mechanism for optical nonlinearity in transparent liquids. Nonlinear refractive index depends linearly on light intensity.

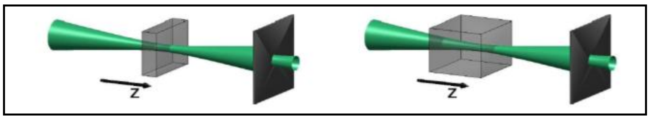

Figure 2: Z-scan: closed aperture geometry.[18]

As in Figure (3) that demonstrates the order of scanning the third dimension of positive and negative refraction of Nonlinear.

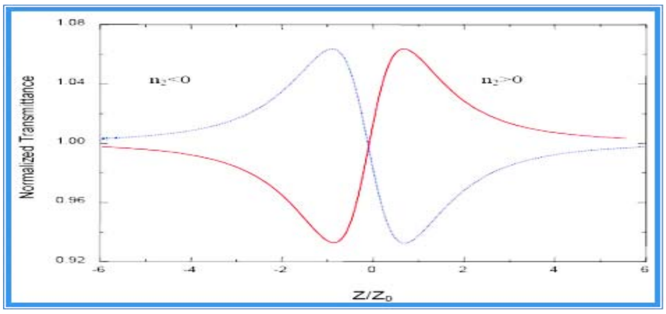

Figure 3: Z-Scan technique –closed aperture Order curves of nonlinear refraction positive and negative[13].

By monitoring the transmittance change through a small circular aperture placed at the far-field position, one is able to determine the nonlinear refractive index. The samples were moved in step of 1 mm during the scan.[13]

The relation between the normalized transmittance T(z) and z position was obtained by moving the samples along the axis of the incident beam (z-direction) with respect to the focal point, We define an easily measurable quantity ΔT_{p-v} as the difference between the normalized peak and valley transmittance: ΔT_{p-v} is linearly-dependent on the temporally averaged induced phase distortion. The variation of this quantity as a function of $\Delta\Phi_o$ is given by : [19]

$$\Delta T_{p-v} = 0.406\Delta\Phi_0 \quad (1)$$

Where $\Delta T_{p-v} = T_p - T_v$ (2)

ΔT_{p-v} the difference in transmittance between the peak and valley. The non- linear refractive index can be obtained from the formula:

$$n_2 = \Delta\Phi_0 / I_0 L_{eff} k \quad (3)$$

$$k = 2\pi/\lambda \quad (4)$$

λ: wavelength of laser beam.

$$I_0 = 2p/\pi w_0^2 \quad (5)$$

I_0: is intensity of the laser beam at the focus (Z = 0).
P: power of laser beam
w_0: the beam radius at the focal point

$$L_{eff} = (1 - exp^{-\alpha_0 t})/\alpha_0 \quad (6)$$

L_{eff} :the effective thickness of the sample, t: is the thickness of the sample, α_0 :the linear absorption coefficient, which can be found from the curved transmission.[20]

$$\alpha_0 = \frac{1}{t} Ln \frac{1}{T} \quad (7)$$

Where t: thickness of sample, T: transmittance.

2.2.2 Open Aperture Z-Scan

Nonlinear absorption of a sample is manifested in the open aperture Z-Scan measurements. For example, if nonlinear absorption like two-photon absorption (TPA) is present, it is manifested in the measurements as a transmission minimum at the focal point. On the other hand, if the sample is a saturable absorber, transmission increases with increase in incident intensity and results in a transmission maximum at the focal region. It has been shown that the model originally developed by Bahae et. al for pure TPA can also be applied to excited state absorption.[17]

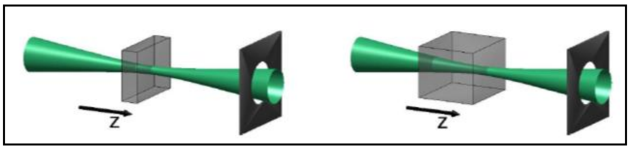

Figure 4: Z-scan: open aperture geometry [18]

The typical geometries for Z-scan measurements are sketched in Fig. (2) and (4). The focused Gaussian beam is propagating in z direction; the crystal is moved through the focus. The integrated intensity will be influenced mainly by the nonlinear absorption, the angular distribution of the intensity, however, will be affected by both nonlinear absorption and refraction. Thus in an open aperture geometry the nonlinear absorption can be measured, in a closed aperture geometry the nonlinear refraction. One has to discriminate whether the sample is thin or thick (compared to z_0 of the Gaussian beams). For both cases comprehensive mathematical descriptions have been developed which can be used for the evaluation of Z-scan measurements.[18]

In Fact, the nonlinear absorption coefficients can be easily calculated from the transmittance curves figure (5) [15].

$$\beta = \frac{2\sqrt{2}}{I_0 L_{eff}} \Delta T \qquad (8)$$

Where ΔT is the one peak value at the open aperture Z-scan curve. The value of β will be positive for saturable absorption and negative for two photon absorption.[20]

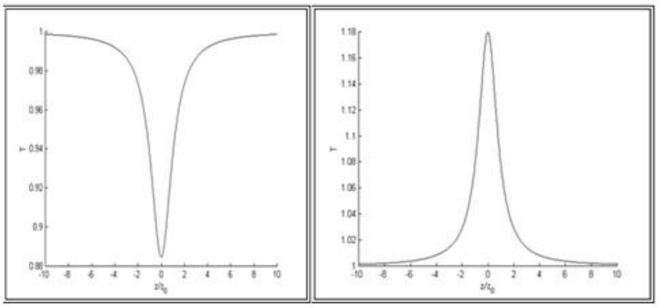

Figure 5: shows Forms Z-Scan technique open aperture.

3. Experimental Setup

To further study the influence of laser intensity on the absorption property, we measured the transmittance of the sample at focal position with different laser intensities. In order to describe the nonlinear optical properties at darkness at room temperature and have high stability. The beam waists in focal plane were measured to be 0.018mm for the wavelengths of 532 nm.

Z-scan measurements were performed in two parts, closed and open aperture, with three input energies. Each case was employed at 1064 nm and 532 nm. The closed-aperture Z-scan was used to measure the nonlinear refractive index, while the open-aperture was used to measure the nonlinear absorption coefficient. At each wavelength the Z-scan experiments were performed at two directions (transmission and reflection).Figure (6) shows the set-up of the Z-scan system.

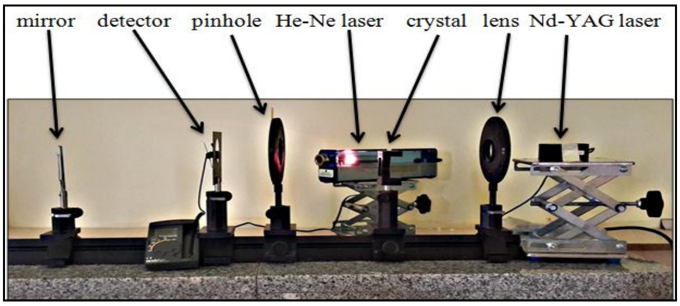

Figure 6: The Setup Of Z- Scan Technique

The He:Ne laser was used for alignment of the optical set-up. The He:Ne laser was placed perpendicular to the setup. Then mirror reflected the laser beam inside the setup. The second element of z-scan system is collimating lens whose focal length is 15 cm, the lens was placed opposite the Nd:YAG laser. The plane mirror was used in alignment, which reflects the laser beam of He:Ne laser backward, and partially transmits laser beam toward to Nd:YAG laser source. The third optical element is the pin hole, which is placed between the sample and nearest to the detector. The diameter of pin hole is 1mm. The fourth optical element in z-scan system is the optical thermal detector, which is used to measure the output power of CW laser. The detector was placed at the far field of a Gaussian laser beam when the He:Ne laser is removed after alignment.

4. Result and Conclusion

The nonlinear optical properties were investigated for Lithium Triborate (LBO) crystal and the thickness of the crystal was (t=5.95mm) with power (P = 25 and 40 mwatt) at each (532 and 1064 nm) of Nd-YAG laser.

4.1 Nonlinear Refractive index

The nonlinear refractive index (n_2) of Lithium Triborate (LBO) crystal was measured by the Z-Scan technique. Figure (7, 8 and 9) shows a Closed–Aperture Z-Scan technique for Lithium Triborate (LBO) crystal at different laser wavelength (532nm and 1064 nm) with different laser powerful (25mwatt, 40mwatt).

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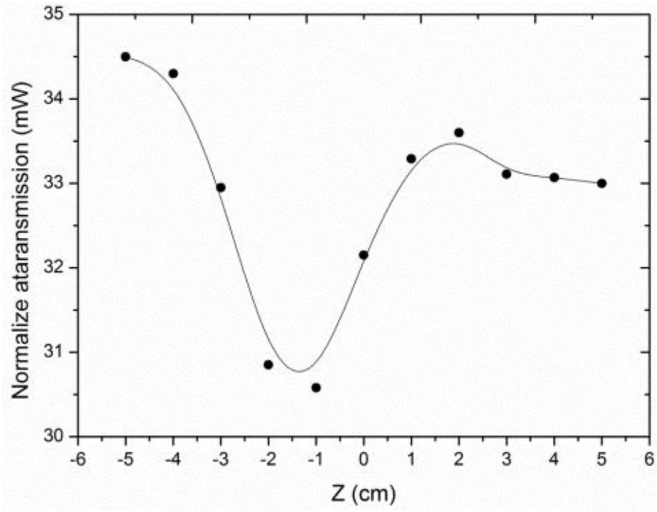

Figure 7: Closed Aperture Z-Scan for LBO in wavelength 532 nm at 40 mW

In figure (7), the nonlinear effect region is extended from (-5)to (5)cm, the valley-peak configuration indicates the positive sign of refractive index nonlinearity ($+n_2$) self-focusing.

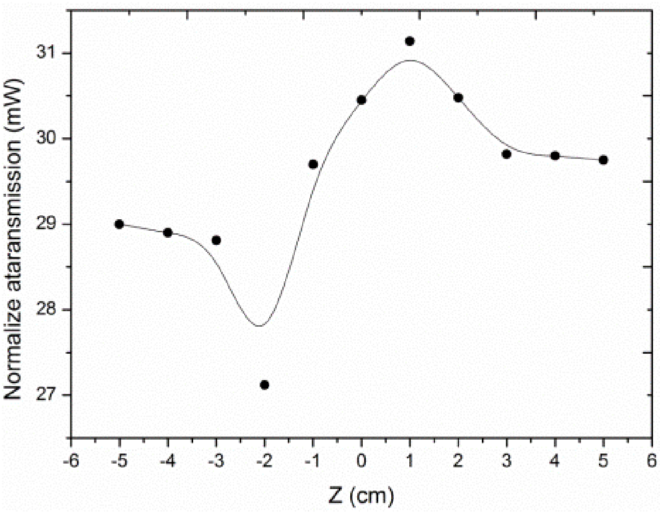

Figure 8: Closed Aperture Z-Scan for LBO in wavelength 532 nm at 25 mW

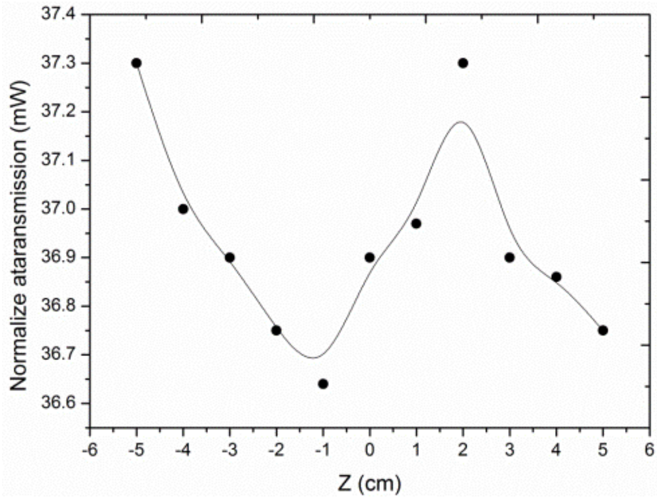

Figure 9: Closed Aperture Z-Scan for LBO in wavelength 1064 nm at 40 mW

Figure (8 and 9) shows the normalized transmittance for the Closed Aperture (CA) curve of LBO. The valley to peak configuration of the curve [Figure (8 and 9)] suggests that the refractive index change is positive, exhibiting a self-focusing effect. This may be an advantage for the application in protection of optical sensors. As seen from the closed aperture Z-scan curve, the prefocal transmittance valley is followed by the post focal peak which is the signature of positive nonlinearity.

4.2 Nonlinear Absorption coefficient

The nonlinear Absorption coefficient (β) of LBO was measured by the Z-Scan technique. Figure 10, 11 and 12 shows Open–Aperture Z-Scan technique for Lithium Triborate (LBO) crystal at different laser wavelength(532nm and 1064nm) with different laser powerful(25mwatt, 40mwatt).

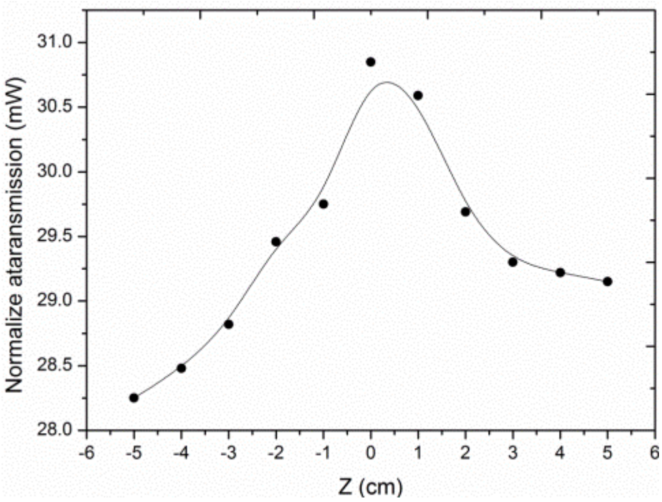

Figure 10: Open aperture Z-Scan for LBO in wavelength 532 nm at 25 mW

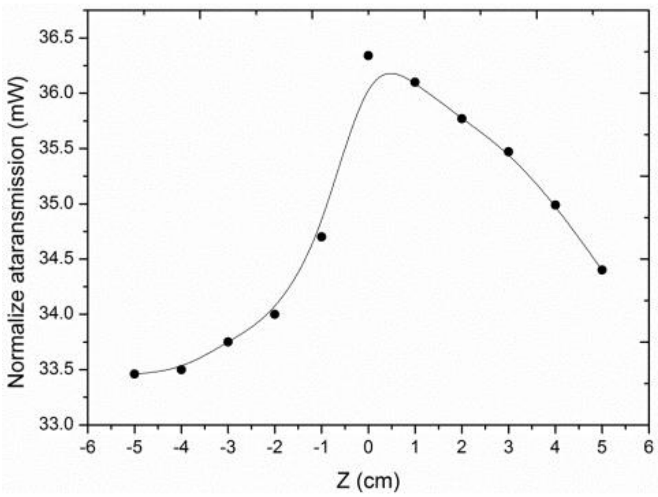

Figure 11: open aperture Z-Scan for LBO in wavelength 532 nm at 40 mW

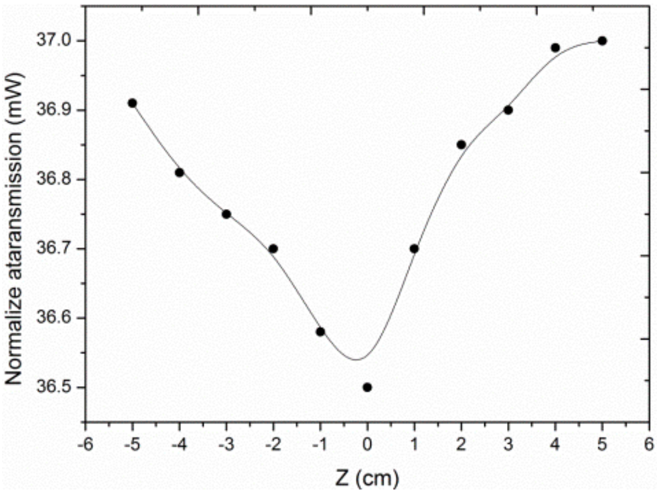

Figure 12: open aperture Z-Scan for LBO in wavelength 1064 nm at 40 mW

Table 2: The results of nonlinear optical properties for Lithium Triborate (LBO) crystal by the Z- scan at different laser powerful (25mwatt and 40mwatt).

$\lambda = 532 \times 10^{-7}$ (cm)		
	Power of laser = 25(mw)	Power of laser = 40(mw)
ΔT_{p-v}	4.02	3.92
I_o (mw/cm^2)	49100	78600
$\Delta\phi_o$ (Rad)	9.9	9.56
n_2 (cm^2/mw)	3.04×10^{-10}	0.18×10^{-10}
T_{max}	30.85	36.34
β (cm/mw)	3.7×10^{-3}	2.3×10^{-3}

$\lambda = 1064 \times 10^{-7}$ (cm)	
	Power of laser = 40(mw)
ΔT_{p-v}	0.66
I_o (mw/cm^2)	78600
$\Delta\phi_o$ (Rad)	1.625
n_2 (cm^2/mw)	6.2×10^{-10}
T_{max}	36.5
β (cm/mw)	2.3×10^{-3}

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