

# Measurements of Nonlinear Absorption and Refraction Coefficients of Pure and Nd Doped Calcium Lanthanum Borate Glasses

Gajanan B. Harde

Department of Physics, Shri R. R. Lahoti Science college, Morshi, Maharashtra, India-444905

E-mail: [gajanangbh\[at\]gmail.com](mailto:gajanangbh[at]gmail.com)

**Abstract:** In the present work, we have calculated optical nonlinear absorption and refraction coefficients of pure and Nd doped calcium lanthanum borate (CLB) glasses prepared by melt quenching method. The measurements were carried out using a single beam z-scan method. Under a linearly polarized continuous wave (cw) He-Ne laser beam at  $0.6328\mu\text{m}$  (red) having 10 mW output power, samples were excited. A closed aperture z-scan method was adopted for nonlinear refractive index ( $n_2$ ) measurement and nonlinear absorption coefficient ( $\beta$ ) was measured by using an open aperture z-scan setup. Study shows the values of  $n_2$  and  $\beta$  for pure and Nd doped CLB glasses are significant.

**Keywords:** Melt quenching method, nonlinear refractive index, absorption coefficients, thermal effects

## 1. Introduction

In last few decades, several nonlinear optical (NLO) materials are developed due to its wide applications such as switching, power limiting, modulation, optical computing, and high optical data storage devices [1-2]. Therefore, it is important to check the nonlinearity of the materials by calculating the values of parameters like  $\beta$  and  $n_2$ . The values of  $\beta$  and  $n_2$  can explain nonlinear behavior and capability of the materials for the desired practical applications [3]. The  $\beta$  and  $n_2$  can estimate using Z-scan technique, which was invented by the Bahae et al. in 1989 [4] for determining various parameters related to the optical nonlinear properties. This is a simple and effective method widely used in material characterization. It can provide magnitudes as well as sign of the nonlinear susceptibility. In this method, an intense laser beam is used to generate nonlinear effect in material. When intense laser beam passes through the sample, its transmittance can change due to nonlinear absorption (NLA) and nonlinear refraction (NLR) of materials. These NLA and NLR of the materials can be estimated by using Z-scan technique in both modes (open aperture and closed aperture, respectively). There are several materials; but glasses are important due to their compositional diversity, different shapes and size, easy fabrication, low cost, and high optical nonlinearities [5-6]. The glasses doped with rare earth ions have attracted much attention of researchers because of their practical applications in the field of lasers, optical devices and sensors, etc [7]. Doping of Rare earth ions can enhance the NLO properties of glasses [8]. There are many papers available in literature about optical properties of rare earth ions doped glasses. The glasses having high optical nonlinearities are good candidates for NLO device applications like signal processing, ultrafast switches and power limiters [9, 10]. The knowledge of NLO properties of materials is an essential to decide the capability of material for the NLO devices.

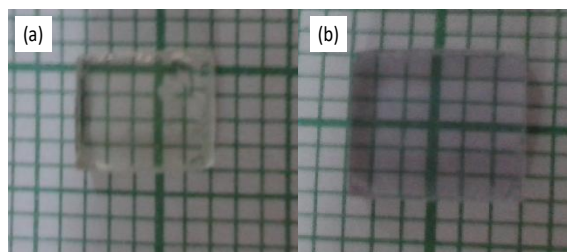
In this research paper, we report the estimated values of  $\beta$  and  $n_2$  of pure and Nd doped calcium lanthanum borate

(CLB) glasses prepared by conventional melt quenching method.

## 2. Experimental

### 2.1 Preparation of Glasses

Polycrystalline powder sample of pure and neodymium (Nd) doped calcium lanthanum borate (CLB) ( $\text{Ca}_3\text{La}_2(\text{BO}_3)_4$ ) were synthesized by using reagents, neodymium oxide ( $\text{Nd}_2\text{O}_3$ ), lanthanum oxide ( $\text{La}_2\text{O}_3$ ), calcium carbonate ( $\text{CaCO}_3$ ) and boric acid ( $\text{H}_3\text{BO}_3$ ) of 99.9% purity as they received without further purification. The pure and 5 mol% Nd<sup>3+</sup> doped glasses with  $\text{La}_2\text{O}-\text{CaO}-\text{H}_3\text{BO}_3$  compositions were obtained from corresponding polycrystalline powder compounds using melt quenching method. Appropriate quantities of polycrystalline powder samples taken in an open silica crucible separately and kept in a handmade high temperature muffle furnace and then the temperature was raised and maintained at 1050 °C for 2 h. The pure and Nd doped CLB glasses were obtained by fast cooling the corresponding melt. The prepared glasses were cut and polished (shown in fig. 1) to use for further studies. Details of the fabrication of these glasses given in the reference [11].



**Figure 1:** Photographs of (a) pure and (b) 5mol% Nd doped CLB glasses.

### 2.2 Method

Volume 12 Issue 4, April 2023

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Figure 2 represent the experimental set up of Z-scan technique used in the present study. We estimated the values of refractive and absorptive non-linearity of the present glasses by performing close and open aperture z-scan technique using linearly polarized cw He-Ne laser beam at wavelength 0.6328 μm (red) with the output power of 10 mW. In this technique, intense laser beam was focused on the sample using a lens; the sample was moved along the beam axis (z-axis) through the focal region over a distance several times of Rayleigh range. Note down the transmitted laser light intensity for each z value and plot graph between transmitted light intensities and z values (Fig. 3 and 4). The transmitted light intensity will be a minimum at the focal point (z = 0). It will increase for other values of z.

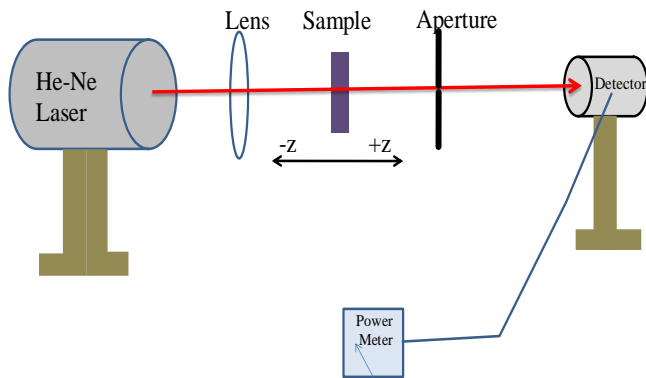


Figure 2: Experimental set up of Z-scan technique

The photon energy of the incident light beam is less than the band gap energy of title glass samples so that  $E_p > E_g/2$ . Hence NLO effects generated in the samples due to two photon absorption (TPA) are expected [12, 13]. In the present study, incident laser light beam was focused on the sample using lens having 10 cm focal length. Then measured the value of beam waist ( $w_0$ ) and the Rayleigh length ( $z_0$ ) were 22 μm and 2.1 mm, respectively. An open aperture Z-scan mode was used in order to estimate the value of intensity dependence nonlinear absorption coefficient of the same samples. The values of  $n_2$  and  $\beta$  were estimated using closed- and open mode of Z-scan technique, which was described by Bahae et al. [4, 14]. The intensity-dependent nonlinear refraction is expressed by the relation.

$$n(I) = n_0 + n_2 I \tag{1}$$

Where,  $n_0$  represent the linear refractive index which can calculate from absorption spectrum,  $n_2$  indicate the NLR index and ‘I’ is the intensity of the laser beam. For a third-order NLO process, nonlinear absorption can be neglected,

### 3. Results

#### 3.1 Z-scan technique for measurement of nonlinear refraction ( $n_2$ ):-

In order to estimate the value of  $n_2$ , thin glass samples of pure and Nd doped CLB glasses were subjected to closed mode of Z scan technique, and value of direct transmittance through aperture was kept constant at  $S = 0.4$ . Well polished sample with 1.5 mm thickness were used in this experiment. The samples were moved along the optic axis (the z-direction) through the focus of a single laser beam. Then the energy transmitted through an aperture was measured by photo detector, which was placed in front of sample as shown in fig. 2. It is convenient to plot the graph between the normalized transmittance of the system and the sample position (z mm). The closed mode Z-scan profiles for glass samples are shown in fig.3. The cw He-Ne laser is able to induce the thermal nonlinear effect in the samples due to high intense. Therefore, the nonlinearity arises in the samples because of the thermal effect produced by the absorption of laser light, which exhibit peak to valley configuration. This configuration represents self-defocusing effect as well as the negative value of  $n_2$ . The value of  $n_2$  can be deduced from equation 4, which is obtained from the relation between phase distortion and difference in transmittance ( $\Delta\Phi_0$  and  $\Delta T_{pv}$ ) [4, 14].

$$\Delta T_{pv} = 0.406(1 - S)^{0.27} \Delta\phi \tag{2}$$

$$\Delta\phi = \frac{2\pi}{\lambda} n_2 I_0 L_{eff} \tag{3}$$

$$n_2 = \frac{\Delta T}{k I_0 L_{eff} 0.406(1 - S)^{0.27}} \tag{4}$$

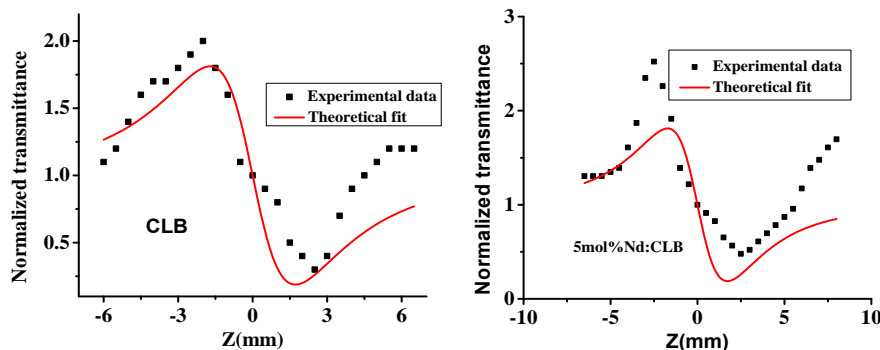


Figure 3: Close mode Z-scan profile of pure and Nd doped CLB glasses.

Where,

$\Delta T_{pv}$ = Difference in transmittance,

$\Delta\Phi_0$ = phase distortion,

S= transmittance without sample through aperture,

$$L_{eff} = \frac{1 - \exp(-\alpha L)}{\alpha}, \text{ effective thickness of sample}$$

( $\alpha$ = linear absorption coefficient and  $L$ = measured thickness of sample) and

$I_0$ = peak intensity at the focus.

The nonlinearity of glass samples can often be evaluated from the difference between the maximum and minimum values of the normalized transmittance ( $\Delta T$ ). By using above expression, the value of  $n_2$  for pure and 5mol% Nd doped CLB glasses were found to be  $-6.5932 \times 10^{-8} \text{ cm}^2/\text{W}$  and  $-7.6075 \times 10^{-8} \text{ cm}^2/\text{W}$  respectively. It shows the value of nonlinear refractive index  $n_2$  is enhanced by doping Nd in CLB glass.

### 3.2 Z-scan technique for measurement of nonlinear absorption ( $\beta$ )

For the estimation of  $\beta$ , aperture as shown in fig.2 was removed from the experiment so that the total beam power or energy could be collecting the photo-detector. Record all transmittance by detector with respect to position of the sample and plot graph (fig.4). Both plots represented valley at the focus indicating the reverse saturable absorption (RSA) [14].

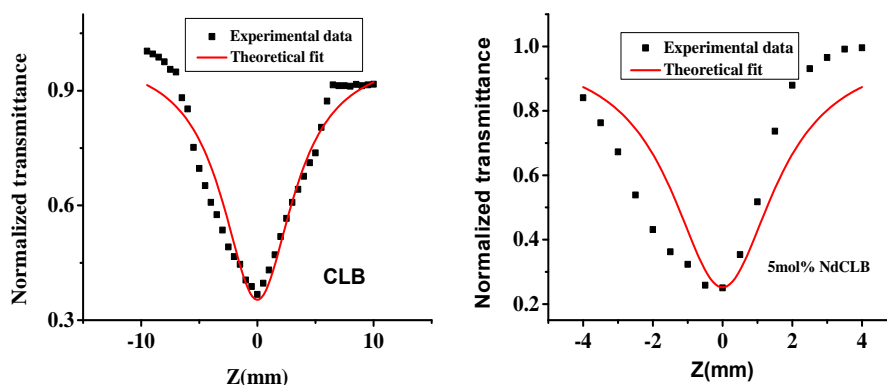


Figure 4: Open aperture Z-Scan profile of pure and Nd doped CLB glasses.

Glasses are finding wide applications in different field such as, in waveguide lasers and amplifiers. But, in isotropic materials (glass) second-order nonlinearities are always absent. The nonlinearities of higher than four orders are also negligible. Therefore, the non-linear transmission fits well to a three-photon type absorption process. From the absorption spectrum of the  $\text{Nd}^{3+}$ -doped glass, it is observed that there is always a strong absorption peak near 581- 675 nm [11]. Hence, using 632.8 nm cw He-Ne laser incidents on the sample, many electrons based on 4 f may transit by doping  $\text{Nd}^{3+}$ , and when returned back to the ground state these electrons emit the photon, resulted in the NLO effect. [15, 16]

The values of  $\beta$  for pure and Nd doped CLB were evaluated from curve fitting and found  $1.39 \times 10^{-6} \text{ cm}/\text{GW}$  and  $1.60 \times 10^{-6} \text{ cm}/\text{GW}$  respectively. From this we can say that, the value of nonlinear absorption coefficient is enhanced with Nd doping in CLB glass.

### 4. Conclusions

In conclusion, pure and 5mol%Nd doped CLB glasses have been fabricated by melt quenching technique. The thin glass samples have been prepared and characterized by Z-scan

It is called an “open aperture” Z-scan because all the transmitted intensity was collected by photo detector when the sample moves through the focus of the Gaussian beam. Glasses having large values of refractive and absorption coefficient are ideal materials for fabricating photonic devices. In borate series glasses, the non-linearity is primarily generate by the  $\text{BO}^{3+}$  ions.

NLA coefficient  $\beta$  can be deduced by using the relation [15, 16].

$$T(z, S=1) = \sum_{m=0}^{\infty} \frac{(-q_0(z))^m}{(m+1)^{3/2}} \text{ for } |q_0(0)| < 1 \quad (5)$$

Where,  $q_0(z) = \frac{\beta I_0 L_{\text{eff}}}{1 - \frac{z^2}{z_0^2}}$ ,  $z_0 = \frac{kw_0^2}{2}$  is represent

Rayleigh's range and  $w_0$  is beam waist.

under linearly polarized cw He-Ne laser beam at  $0.6328 \mu\text{m}$  (red) with output power 10 mW, and confirmed increase in values of  $n_2$  and  $\beta$ .

### Acknowledgements

Authors GGM, GBH acknowledge for financial support from University Grant Commission, under major research project (F. No. 41-925/2012 (SR)) and minor research project (F. No. 47-1363/10 (WRO)) respectively.

### References

- [1] Y. Lin, J. Zhang, L. Brozowski, E. H. Sargent and E. Kumacheva, J. Appl. Phys. 91(2002)522.
- [2] F. Li, Q. G. Zheng, N. L. Dai, R. X. Lu, Mater. Lett. 62 (2008)3095.
- [3] S. L. Mathews, S. Chaitanya Kumar, L. Giribabu, S. Venugopal Rao, Mater. Lett. 61(2007)4426.
- [4] M. Sheik-Bahae, A.A. Said, E.W. Van Stryland, High-sensitivity, single-beam  $n_2$  measurements, Opt. Lett. 14, (1989)955.
- [5] J. M. P. Almeida, L. De Boni, A. C. Hernandez, C. R. Mendoca, Opt. Express 19 (2011)17220.

- [6] K. Terashima, S. Tamura, S. H. Kim, T. Yoko, J. Am. Ceram. Soc. 80 (1997) 2903.
- [7] S. Arunkumar, K. Marimuthu, J. Alloys Compd. 565 (2013) 104.
- [8] Y. Cheng, H. Xiao, W. Guo, Ceramics International 34 (2008) 1335.
- [9] R. C. Beltran, H. Desirena, G. R. Ortiz, E. D. L. Rosa, G. Lanty, J. S. Lauret, S. R. Servin, A. Schulzgen, J. Appl. Phys., 110 (2011) 083110.
- [10] R. L. Thomas, Vasuja, M. Hari, V. P. N. Nampoori, P. Radhakrishnan, S. Thomas, J. Opto.Elec. Adv. Mater., 13 (2011) 523.
- [11] Gajanan B. Harde and Gajanan G. Muley, Study of Spectroscopic Properties of Pure and Nd Doped  $\text{Ca}_3\text{La}_2(\text{BO}_3)_4$  Glasses, Procedia Technology 24, (2016) 727 – 732
- [12] B. Karthikeyan, Reji Philip, S. Mohan, Optical and non-linear optical properties of  $\text{Nd}^{3+}$ -doped heavy metal borate glasses, Optics Communications 246, (2005) 153.
- [13] Vinay Kumari, Vinod Kumar, B.P. Malik, R.M. Mehra, Devendra Mohan, Opt. Commun. 285, (2012) 2182.
- [14] M. Sheik-Bahae, A.A. Said, T.H. Wei, D.J. Hagan, E.W. Van Stryland, IEEE J. Quantum Elect. 26, (1990) 760.
- [15] K.K. Nagaraja, S. Pramodini, A. Santhosh Kumar, H.S. Nagaraja, P. Poornesh, DhananjayaKekuda, Opt. Mater. 35, (2013) 431.
- [16] Y. S. Tamgadge, V. G. Pahunkar, S. S. Talwatkar, A. L. Sunatkari, G. G. Muley, Thermally stimulated third-order optical nonlinearity in Cd-doped CuO–PVA thin films under cw laser illumination, Appl. Phys. B, (2015)