Study on the 3rd Order Nonlinear Optical Properties of LaCOB and Nd:LaCOB glasses

Madhav Namdev Rode
Department of Physics, Vaidyanath College, Parli - Vaijnath, Dist. Beed (M.S.) India

Abstract- **The glasses of system (x)Nd_2O_3-(1-x)La_2O_3-8CaO-3B_2O_3 with x=0 (LaCOB) and 0.03 (Nd:LaCOB) have been synthesized by melt quench technique. 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 µm (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 (β) was measure by using an open aperture z-scan setup. Study shows the values of n_2, β and susceptibility ($\chi^{(3)}$) for LaCOB and 3% Nd doped LaCOB glasses are significant.**

Keywords: **Powder X-ray diffraction; nonlinear optical properties, Nonlinear absorption coefficient, Z-scan, He-Ne laser.**

I. INTRODUCTION

In last few decades, nonlinear optical materials are taking much attraction due to its wide applications such as switching, power limiting, modulation, optical computing, and high optical data storage devices [1-3]. Therefore, it is important to check the nonlinearity of the materials by calculating the values of parameters like optical absorption coefficient (β), refractive index (n_2) and susceptibility ($\chi^{(3)}$). The values of β and n_2 can explain nonlinear behavior and capability of the materials for the desired practical applications [4]. The β, n_2 and ($\chi^{(3)}$) can estimate using Z-scan technique, which was invented by the Bahae et al. in 1989 [5, 6] for determining various parameters related to the optical nonlinear properties. Different configurations can be implemented, in particular the open aperture and closed-aperture [7]. This is a simple and effective method widely used in material characterization. It can provide magnitudes as well as sign of the nonlinear susceptibility $\chi^{(3)}$ [8]. Doping of Rare earth ions can enhance the NLO properties of glasses [9]. There are many papers available in literature about the glasses having high optical nonlinearities are good candidates for NLO device applications like signal processing, ultrafast switches and power limiters [10-12]. The knowledge of NLO properties of materials is an essential to decide the capability of material for the NLO devices.

We have already reported the preliminary study on (x)Nd_2O_3-(1-x)La_2O_3-8CaO-3B_2O_3 with x = 0 (LaCOB) and 0.03 (Nd: LaCOB) glass was successfully grown using a simple melt quenching technique. In this report we were discussed on results of X-ray diffraction, UV-VIS and FTIR of material. Moreover the parameters such as optical transparency and energy band gap also have been discussed [13].

In this research paper, we report the estimated values of β, n_2 and susceptibility of pure and Nd doped lanthanum calcium borate (LaCOB) glasses prepared by simple melt quenching method.

II. EXPERIMENTAL

Material and methods

A Lanthanum oxide (La_2O_3) was purchased from Central Drug House (CDH) (P) Ltd, India. Calcium carbonate ($CaCO_3$) was purchased from Fisher scientific, India. Neodymium oxide (Nd_2O_3) was purchased from LOBA Chemie, India. Boric acid (H_3BO_3) were purchased from sd Fine chemicals, Mumbai, all these chemicals used were of analytical reagent grade.

An alkali borate glasses (x)Nd_2O_3-(1-x)La_2O_3-8CaO-3B_2O_3 with x = 0 and 0.03 were prepared by the melt-quenching method. The chemicals La_2O_3, $CaCO_3$, Nd_2O_3 and H_3BO_3 with 99.99% purity used as the starting raw material. Nd_2O_3 was mixed x = 0 and 0.03 with La_2O_3, $CaCO_3$, and H_3BO_3. Appropriate amounts of the raw materials were crushed in a mortar with the help of pestle to make the homogeneous mixture. A detail about the temperature variation and heating time for synthesis of glasses was given in the reference [13].

III. RESULT AND DICUSSION

Third order Nonlinearity Properties

Bahae et al. reported the most crucial and sensitive Z-scan technique to evaluate the nonlinearities originating in a material when irradiated with highly repetitive optical energy [5].

The most important parameter in the study of nonlinearity is third order nonlinear optical susceptibility which is responsible for phenomena like third harmonic generation. The optical resolution of Z-scan setup is tabulated in table 1.

Table 1: Optical resolution of Z-scan setup

Parameters	Magnitude
Laser wavelength (λ)	632.8 nm
Laser power (P)	10 mW
Lens focal length (f)	20 cm
Optical path distance (Z)	113 cm
Beam waist radius (w_a)	20μm
Aperture radius (r_a)	1.5 mm
Incident intensity at the focus (I_o)	2.3375 mW/m^2

Firstly the sample was placed at the focus (Z = 0) of the beam irradiated path. After that the sample was smoothly shifted in +Z and Z direction with reference to focus. The variations in intensity of nonlinear refraction attributed by LaCOB and Nd:LaCOB glasses has been detected and recorded using the photo detector placed at far field. The closed aperture Z-scan transmittance curve of LaCOB and Nd:LaCOB glasses is shown in Fig. 1a and 2c. The result shows that the glasses medium offers phase shift in propagating light from valley to peak (positive refraction) due to the intrinsic property of material exhibiting self-focusing nature [14]. The phase shift in direction of propagating light evidences the origin of nonlinear refraction which is attributed by the special distribution of energy along the glass surface due to incident laser beam giving rise to localized thermal lensing effect [15].

The on axis phase shift (D/) in terms of peak to valley transmission (ΔTp-v) is given as [4],

$$\Delta Tp\text{-}v = 0.406\ (1-S)^{0.25} l \Delta\phi l \qquad (1)$$

Where $S = [1-\exp(-2r_a^2/w_a^2)]$ is the aperture linear transmittance, r_a is the aperture radius and w_a is the beam waist radius in front of aperture.

The nonlinear refraction (n_2) of LaCOB and Nd:LaCOB glasses has been determined using the relation [5],

$$n_2 = \Delta\phi / K I_o L_{eff} \qquad (2)$$

where $K = 2\pi/\lambda$, $I_0 = 2P/\pi wa^2$, is the incident irradiance intensity of beam at the focus (Z = 0).

The effective thickness of the sample has been determined using the equation,

$$L_{eff} = [1 \exp(-\alpha L)]/a$$

where α is the linear absorption coefficient and L is the thickness of the sample.

The nonlinear refraction (n_2) of LaCOB and Nd:LaCOB glasses is found to be of order of 10^{-9} cm^2 /W.The high magnitude of positive nonlinear refraction of prepared glasses shows the potential kerr lens modelocking ability which makes it liable for utility in shorter pulse generation and laser alignment systems [14, 15].

By employing the open aperture Z-scan technique, the signature and magnitude of nonlinear absorption coefficient (β) of LaCOB and Nd: LaCOB glasses have been determined. The open aperture Z-scan transmittance curve of LaCOB and Nd: LaCOB glasses is shown in Fig. 1b) and 2d) which evidences that the transmittance of LaCOB and Nd:LaCOB glasses falls to minimum value as the sample approaches the focus position.

The fall of transmittance with respective to sample position at the focus is termed as reverse saturable absorption effect and such phenomenonfound in LaCOB and Nd:LaCOB glasses might have been contributed by complex fact such as multiphoton absorption assisted by excited state absorption [16]. The material exhibiting reverse saturable absorption effect seeks huge advantage in laser assisted optical switching devices [17-20]. The magnitude of third order nonlinear optical absorption coefficient (β) has been evaluated using the relation [5],

$$\beta = \frac{\Delta T 2\sqrt{2}}{IoLeff} \qquad (3)$$

where ΔT is the one valley value obtained in open aperture Z-scan curve.

The magnitude of β of LaCOB and Nd:LaCOB glass is found to be order of 10^{-4} cm/W. The polarizing nature of glass has been confirmed from the high magnitude of third order nonlinear susceptibility ($\chi^{(3)}$) which has been evaluated by solving the equations [5],

$$Re\chi^{(3)}(esu) = 10^{-4}\,(\varepsilon_0 C^2 n_o^2\, n_2)/(cm^2/W) \qquad (4)$$

$$Im\,\chi^{(3)}(esu) = 10^{-2}\,(\varepsilon_0 C^2 n_o^2\, \lambda\beta)/(cm^2/W) \qquad (5)$$

$$\chi^{(3)} = \sqrt{(Re\chi^{(3)})^2 + (Im\chi^{(3)})^2} \qquad (6)$$

Where ε_0 is the vacuum permittivity, n_0 is the linear refractive index of the sample and C is the velocity of light in vacuum.

The third order nonlinear optical susceptibility of LaCOB and Nd: LaCOB glass is found to be 8.804 and 9.515 esu respectively. The enhanced delocalization of π-electron along the extended network of glass complex due to illumination of high intensity laser beam is the key factor responsible for high magnitude of cubic susceptibility [8].

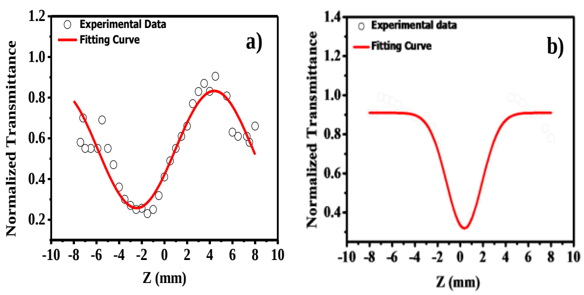

Fig. 1 Z-scan transmittance curve with a) LaCOB close and b) LaCOB open aperture.

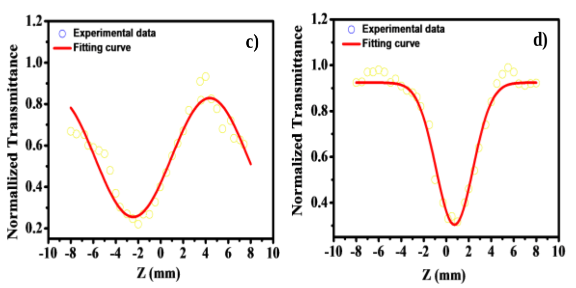

Fig. 2 Z-scan transmittance curve with c) close and d) 3% Nd:LaCOB open aperture.

Table 2 The values of β, NLO refractive index (n_2) and imaginary part of susceptibility ($\chi^{(3)}$)for LaCOB and Nd doped LaCOB samples are tabulated as shown in table 2.

Glass	NLO Refractive Index n_2 (cm^2/W)	β (cm/ W)	Real χ	Imaginary χ	χ^3 (esu)
LaCOB	3.08 X 10^{-9}	4.2 X 10^{-4}	1.59 x 10^{-5}	1.12 X10^{-3}	8.804
3% Nd doped LaCOB	2.85 X 10^{-9}	4.2 X 10^{-4}	1.47 x 10^{-5}	1.12 X10^{-3}	9.515

IV. CONCLUSION

In conclusion, LaCOB and 3% Nd doped LaCOB glasses have been prepared by melt quenching technique. Z-scan analysis confirmed the existence of promising third order nonlinear optical properties in LaCOB and Nd:LaCOB glasses. The doped glass is found to exhibit the positive refraction nonlinearity (self-focusing nature) at 632.8 nm. The n_2 with high magnitude of LaCOB and Nd:LaCOB are 3.08 X10^{-9} 2.85 X10^{-9} cm^2/W. The multiphoton absorption assisted by excited state absorption favored the reverse saturable absorption in LaCOB and Nd:LaCOB glasses with the β of magnitude 4.2 X10^{-4} cm/W. The cubic susceptibility of order 8.804 and 9.515 esu confirmed the high polarizing nature. This property is very interesting for nonlinear applications.

ACKNOWLEDGMENT

Author (Madhav N Rode) is grateful to the Prof. G. G. Muley , HOD, Department of Physics, Sant Gadge Baba Amravati University, Amravati, Maharashtra, India for providing the Experimental setup and characterization THG, SHG facilities and for their help, support, and encouragement.

REFERENCES

[1] Y. Lin, J. Zhang, L. Brozowski, E. H. Sargent and E. Kumacheva, J. Appl. Phys. 91 (2002) 522.

[2] G. B. Harde and G. G. Muley, Spectroscopic and nonlinear optical studies of pure and Nd-doped lanthanum strontium borate glasses, AIP Conf. Proc., 1728, 020405-1-4 (2016).

[3] F. Li, Q. G. Zheng, N. L. Dai, R. X. Lu, Mater. Lett. 62 (2008)3095.

[4] S. L. Mathews, S. Chaitanya Kumar, L. Giribabu, S. VenugopalRao, Mater. Lett. 61 (2007) 4426.

[5] M. Sheik-Bahae, A.A. Said, E.W. Van Stryland, High-sensitivity, single-beam n_2 measurements, Opt. Lett. 14, (1989) 955.

[6] M. Sheik-bahae, A.A. Said, E.W. Van Stryland, Opt. Lett. 14 (1989) 955–957

[7] C. Wiechers, Journal of the Optical Society of America B Vol. 36, Issue 1, pp. 61-68 (2019).

[8] N. Sugimoto, H. Kanbara, S. Fujiwara, K. Tanaka, Y. Shimizugawa, and K. Hirao, J. Opt. Soc. Am. B 16, 1904 1999; T. Hasegawa, T. Nagashima, and N. Sugimoto, Opt. Commun. 250, 411 2005.

[9] Y. Cheng, H. Xiao, W. Guo, Ceramics International 34 (2008) 1335.

[10] S. R. Dagdale, G. B. Harde, V. G. Pahurkar and G. G. Muley, "Synthesis and optical properties of borate glass of system $3Li_2O$-$2K_2O$-$5B_2O_3$", *ISCA-Research Journal of Chemical Sciences*, Res. J. Chem. Sci., Vol. 7(3), 1-3, March (2017).

[11] 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.

[12] R. L. Thomas, Vasuja, M. Hari, V. P. N. Nampoori, P. Radhakrishnan, S. Thomas, J. Opto.Elec. Adv. Mater., 13 (2011) 523.

[13] S. R. Dagdale, G. G. Muley, "Optical study of neodymium doped lanthanum calcium borate glasses of La_2O_3-8CaO-$3B_2O_3$ system", *Bionano frontier*, Vol. 8(3), pp. 265-268 (2015).

[14] A.S. L. Gomes et-al, Third-order nonlinear optical properties of bismuth-borate glasses measured by conventional and thermally managed eclipse Z scan Journal of applied physics, 101, 033115 (2007)

[15] S.M. Lima, J.A. Sampaio, T. Catunda, A.C. Bento, L.C.M. Miranda, M.L. Baesso, J. Non-Cryst. Solids 273 (2000) 215–227.

[16] A. Jha, B. Richards, G. Jose, T. Teddy-Fernandez, P. Joshi, X. Jiang, J. Lousteau, Prog. Mater. Sci. 57 (2012) 1426–1491.

[17] S.M. Lima, T. Catunda, Phys. Rev. Lett. 99 (2007) (243902-4).

[18] J. M. Harbold, F. Ö. Ilday, F. W. Wise, J. S. Sanghera, V. Q. Nguyen, L. B. Shaw, and I. D. Aggarwal, Opt. Lett. 27, 119 2002.

[19] T. Godin, R. Moncorgé, J.L. Doualan, M. Fromager, K. Ait-Ameur, R.A. Cruz, T. Catunda, J. Opt. Soc. Am. B 29 (2012) 1055–1064.

[20] T. Godin, R. Moncorgé, J.L. Doualan, M. Fromager, K. Ait-Ameur, R.A. Cruz, T. Catunda, J. Opt. Soc. Am. B 29 (2012) 1055–1064.