



## ECLIPSING Z-SCAN MEASUREMENT FOR MIXTURE OF DYES (R6G, RB, AND RC) IN LIQUID AND SOLID MEDIA

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### ABSTRACT

This research is a study of the nonlinear optical properties for liquid and solid sample (dye doped polymer) at different concentration in Chloroform solvent by using a high sensitive method known as EZ-Scan technique. EZ-scan experiment was performed using Nd-YAG (SHG) CW laser at 532(nm) in two parts. The first part by using a disk placed in front of the detector to measure the nonlinear refractive index and in the second part removing the disk (open aperture) to measure the nonlinear absorption coefficient and then to measure third order susceptibility. The results suggest that the solid samples were appeared clearer than liquid samples since the nonlinear refractive index is found to be of the order of  $10^{-11}(\text{cm}^2/\text{mw})$ . The magnitude of nonlinear susceptibility is of the order of  $10^{-4}[\text{cm}/\text{watt}]$ . The nonlinear absorption coefficient is of the order of  $10^{-4}(\text{cm}/\text{mw})$ .

**Keywords:** EZ-scan technique, Xanthenes dye, Rhodamine B, Rhodamine C, Rhodamine 6G, nonlinear optics, nonlinear refractive index.

### 1. INTRODUCTION

Dye laser is the most important and most versatile since it is one of the most successful laser sources due to their important contribution to science and technology [1] and has many applications in optics, electronics, non-linear optics, dyes chemistry, local area communication networks, and sensors [2-5]. To overcome the problems related to the nature of liquid systems, solid matrices containing host doped with laser dyes have been developed with the aim of developing a practical solid state dye laser. The most commonly used polymeric material is poly methyl methacrylate (PMMA) because of its best optical transparency for pumping wavelength and its good resistance to laser damage [6]. A review of literature shows that most of the work (spectral and nonlinear) on dye has been done with Rhodamine dye [7-15]. Organic dyes can show very high nonlinear coefficients, due to these compounds can show large variety at high intensities [16]. Nonlinear optics is the study of phenomena that happen as a result of the amendment of the optical properties of a material system by the presence of light. Typically, the prefer light to modify the optical properties of a material system is laser due to high intensity [17]. As the Z-scan method which depends on propagation of a phase distortion to produce a transmittance change since the minimum detectable signal is determined by how small change in transmittance can be measured. The interferometric sensitivity derived from the interference of different parts of the spatial profile in the far field. Recently, it was clear that by looking at the outer edges of the beam in the far field rather than the central part as in the Z-scan, the sensitivity could be significantly increased. This is achieved by replacing the apertures as in Z-scan with disks which works to prevent the central part of the beam. The light that seeps around the edges which appears as an eclipse, thus the name EZ-scan for eclipsing

Z-scan [18]. As with the Z scan, a sample is scanned along the Z axis of a focused beam. In the case of self-focusing ( $n_2 > 0$ ) the sample will behave as a positive lens near the focus. Thus, for the sample positioned before the focus, the far-field beam divergence is increased, and the transmittance will increase by the disk in the far field this is precisely opposite the decrease of transmittance by the aperture for a Z scan. With the sample positioned after the focus, the sample will collimate the beam, and the disk will block more of the light. With the sample positioned after the focus, the sample will collimate the beam, and the disk will block more of the light. Thus, in the EZ scan a self-focusing medium causes an increase in transmittance (peak), followed by a decrease (valley) when the sample is scanned from in front of to behind the focus. For self-defocusing media, the positions of the valley and peak are reversed [19]. As with the Z scan, both NLR and NLA can be study by performing successive EZ scans with and without the disk. An open-aperture Z scan that is sensitive only to nonlinear losses is done by removing the disk [20].

### 2. EXPERIMENTAL WORK

#### 2.1 Materials and instrument

Three well known groups of Rhodamine dyes laser which belong to xanthene family and the molecular weight for these dyes about 479.02 [gm/mole], such as R6G the molecular formula ( $\text{C}_{28}\text{H}_{31}\text{N}_2\text{O}_3\text{Cl}$ ), RC its scientific name (Diethyl-m-amino phenolphthalein) and molecular formula ( $\text{C}_{28}\text{H}_{31}\text{O}_3\text{N}_2\text{Cl}$ ). RB, its scientific name (Tetraethylrodamine) with molecular formula ( $\text{C}_{28}\text{H}_{31}\text{N}_2\text{O}_3\text{Cl}$ ). These dyes were supplied from (HIMEDIA) India Company. These three laser dyes were dissolved in chloroform solvent whose scientific name (Trichloro Methane) with a chemical formula  $\text{CHCl}_3$  and



molecular weight ( $M_w$ ) about 119.38 [g/mole], and refractive index (1.4459), and Productive company Analar company (England) at different concentrations ( $1 \times 10^{-5}$ ,  $2 \times 10^{-5}$ ,  $5 \times 10^{-5}$ ,  $7 \times 10^{-5}$ , and  $1 \times 10^{-4}$ ) [mole/L]. All five concentration are doped in polymer at room temperature by (casting method) to produce thin films. The polymer that used in this research is (PMMA) which its refractive index about (1.49), molecular weight of (PMMA) about 84000 [g/mole], supplied from the company (ICI) German. Sample is moving along the z-axis through the focal point. The transmittance is measured and plotted as a function of sample position along the z-axis, with the focus of the laser beam chosen to be at  $z = 0$ . EZ-scan experiments were performed using a 532 [nm] Nd-YAG (SHG) CW laser beam supplied from (Changchun) company focused by a lens of 12 [cm] focal length. The laser beam waist  $w_0$  at the focus was measured to be 0.0675 [mm] and the Rayleigh length  $Z_R$  was 26.9 [mm] (where  $Z_R$  is the diffraction length of the laserbeam, ( $Z_R = K w_0^2 / 2$ )). The schematic of the experimental setup used is shown in Figure-1.

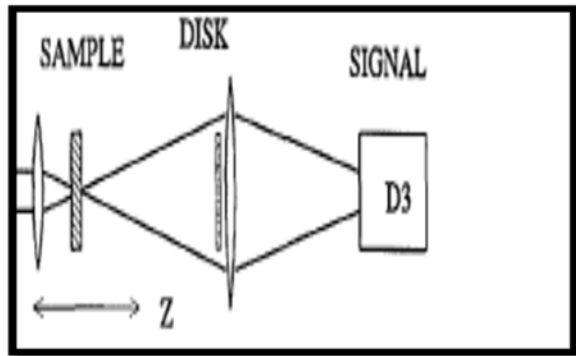


Figure-1. EZ-Scan system.

Photo detector is used to measure the transmission of the beam through an aperture placed in the far field by using operating at wavelength(400-1100) and it is supplied from (Changchun) company and its type is (S121C), the rate of energy (1-500) [mw]. A typical closed aperture EZ-scan curve and open aperture EZ-scan of the dye solution for concentration ( $1 \times 10^{-5}$ ,  $2 \times 10^{-5}$ ,  $5 \times 10^{-5}$ ,  $7 \times 10^{-5}$ , and  $1 \times 10^{-4}$ ) [mole/L] exhibiting the normalized transmittance is shown in the Figure-2 and Figure-3 respectively at incident intensity  $I_0 = 1.39$  [KW/cm<sup>2</sup>]. The curves are characterized by a pre-focal peak followed by a post-focal valley for which implies that the nonlinear refractive index is positive ( $n_2 > 0$ ) for concentration while the transmittance profile at open-aperture (OA) EZ-scan experiment shows (Two Photon Absorption). This may be attributed to the fact that the number of the dye molecules increases when the concentration increases and more particles get thermally agitated resulting in an enhanced effect. The nonlinear refractive index ( $n_2$ ) and nonlinear absorption coefficient ( $\beta$ ) values of the dye solution are

given in Table-1. The corresponding  $\chi^{(3)}$  values calculated from data of nonlinear refractive index and nonlinear absorption coefficient. The nonlinear relationship of refractive index, absorption coefficient, and nonlinear susceptibility shown in Figure-4. A typical closed aperture EZ-scan curve and open aperture EZ of thin films for concentration ( $1 \times 10^{-5}$ ,  $2 \times 10^{-5}$ ,  $5 \times 10^{-5}$ ,  $7 \times 10^{-5}$ , and  $1 \times 10^{-4}$ ) [mole/L] exhibiting the normalized transmittance is shown in the Figure-5 and Figure-6 respectively. The curves are characterized by a post-focal valley followed by a pre-focal peak for which implies that the nonlinear refractive index is negative ( $n_2 < 0$ ) for concentration while the transmittance profile at open-aperture (OA) EZ-scan experiment shows (Saturation absorption) and this occurs because of a change in the intensity of the laser travel through the beam waist in the case of high intensity incident on the sample. The behaviour of the saturation absorption is due to the aggregation of molecules in the singlet excited state leading to depletion of the ground state, which caused a decrease in the intensity at the focus and exhibited high transmittance. The nonlinear refractive index ( $n_2$ ) and nonlinear absorption coefficient ( $\beta$ ) values of the thin films are given in Table-2. The nonlinear relationship of refractive index, absorption coefficient, and nonlinear susceptibility shown in Figure-7.

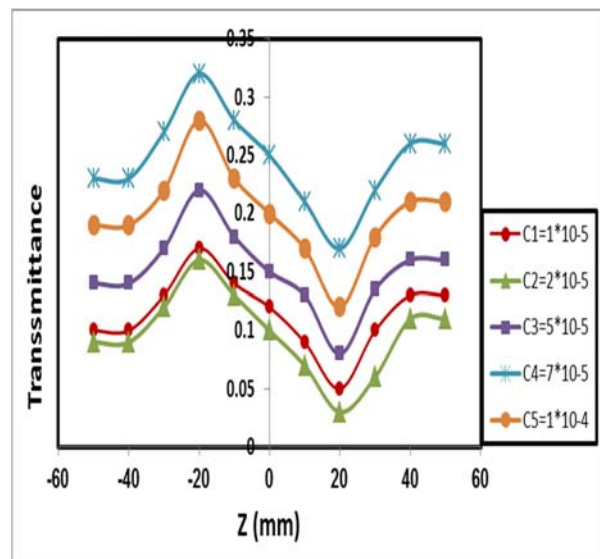


Figure-2. Closed aperture EZ scan data for different concentration of dye solution at  $I_0 = 1.39$  [KW/cm<sup>2</sup>] and ( $S=0.91$ ).

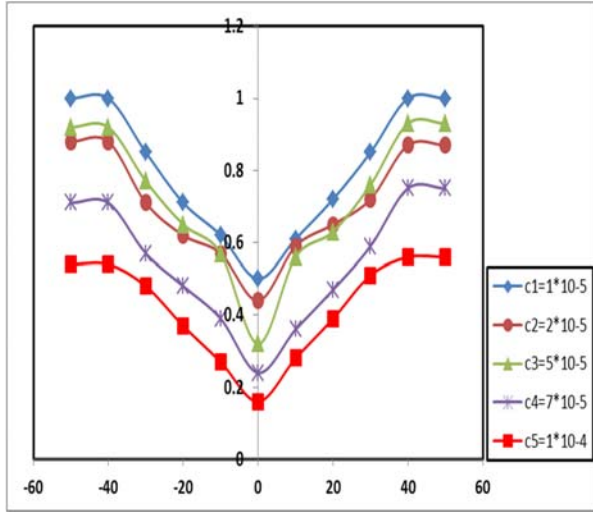
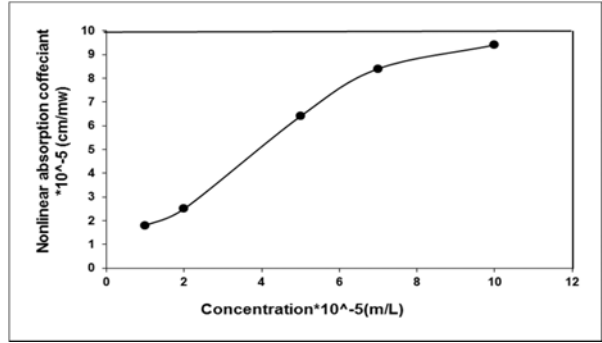
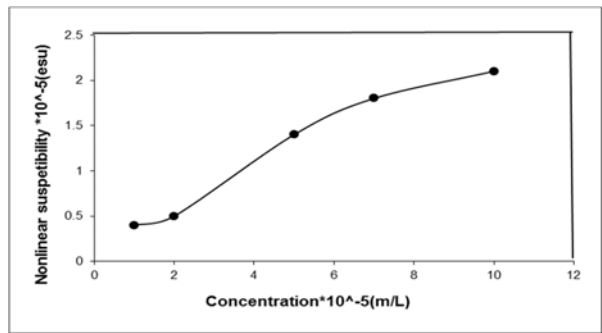


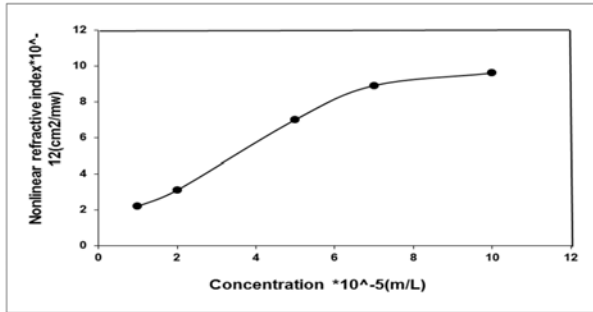
Figure-3. Open aperture EZ scan for dye solutions at  $I_0 = 1.39[\text{KW}/\text{cm}^2]$  and  $(S=1)$ .



(B)



(C)



(A)

Figure-4. Nonlinear relationship for dye solutions (A) Refractive index, (B) Absorption coefficient, (C) Nonlinear susceptibility.

Table-1. Shows nonlinear parameters for dye solution.

| C mole/L           | T%    | $\alpha$ $\text{cm}^{-1}$ | $n_0$  | $L_{\text{eff}}$ cm | $\Delta T_{\text{pv}}$ | $\Delta\Phi_0$ | $n_2$ $\text{cm}^2/\text{mw}$ | T (z) | $\beta$ $\text{cm}/\text{mw}$ | $\chi^{(3)}$ esu     |
|--------------------|-------|---------------------------|--------|---------------------|------------------------|----------------|-------------------------------|-------|-------------------------------|----------------------|
| $1 \times 10^{-5}$ | 30.01 | 12                        | 1.4458 | 0.0582              | 0.12                   | 0.0611         | $2.2 \times 10^{-12}$         | 0.97  | $1.8 \times 10^{-5}$          | $4 \times 10^{-6}$   |
| $2 \times 10^{-5}$ | 15.33 | 18.7                      | 1.4459 | 0.0452              | 0.13                   | 0.0662         | $3.1 \times 10^{-12}$         | 0.44  | $2.5 \times 10^{-5}$          | $5.6 \times 10^{-6}$ |
| $5 \times 10^{-5}$ | 1.06  | 46                        | 1.4460 | 0.0215              | 0.14                   | 0.0713         | $7 \times 10^{-12}$           | 0.32  | $6.4 \times 10^{-5}$          | $1.4 \times 10^{-5}$ |
| $7 \times 10^{-5}$ | 0.41  | 54.9                      | 1.4465 | 0.0181              | 0.15                   | 0.0764         | $8.9 \times 10^{-12}$         | 0.24  | $8.4 \times 10^{-5}$          | $1.8 \times 10^{-5}$ |
| $1 \times 10^{-4}$ | 0.39  | 55.4                      | 1.4467 | 0.0179              | 0.16                   | 0.0815         | $9.6 \times 10^{-12}$         | 0.16  | $9.4 \times 10^{-5}$          | $2.1 \times 10^{-5}$ |

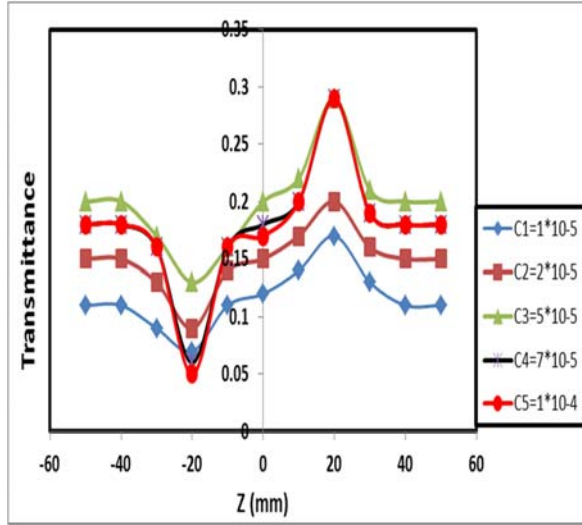


Figure-5. Closed aperture EZ scan data for thin films at  $I_0 = 1.39[\text{KW}/\text{cm}^2]$  and  $(S=0.91)$ .

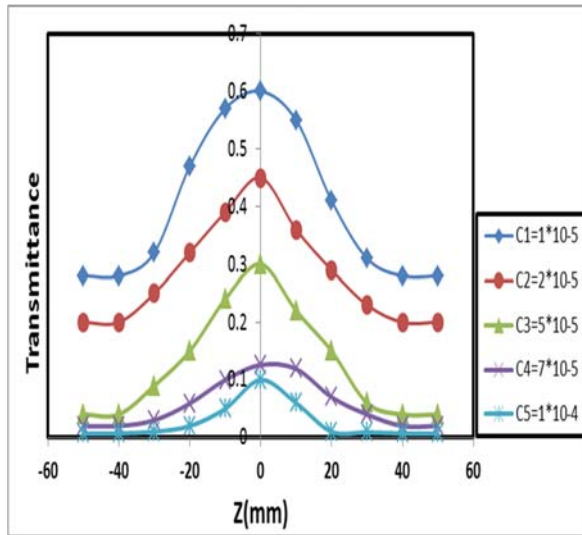
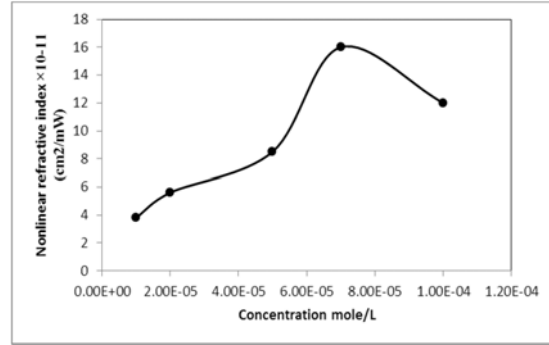
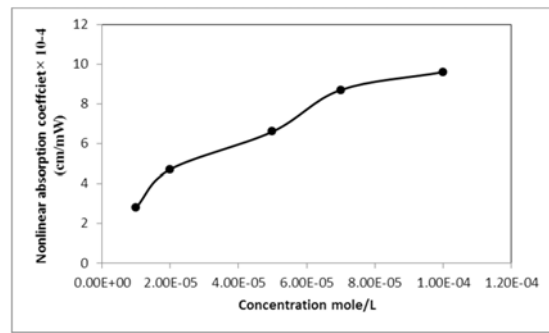


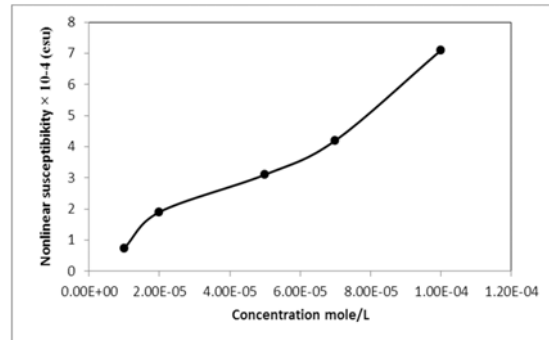
Figure-6. Open aperture EZ scan for thin films at  $I_0 = 1.39[\text{KW}/\text{cm}^2]$  and  $(S=1)$ .



(A)



(B)



(C)

Figure-7. Nonlinear relationship for thin films (A) Refractive index, (B) Absorption coefficient, (C) Nonlinear susceptibility.

Table-2. Shows nonlinear parameters for thin films.

| C mole/L           | T%    | $\alpha$ $\text{cm}^{-1}$ | $n_0$ | $L_{\text{eff}} \times 10^{-3} \text{ cm}$ | $\Delta T_{\text{pv}}$ | $\Delta \Phi_0$ | $n_2$ $\text{cm}^2/\text{mW}$ | T(z)  | $\beta$ $\text{cm}/\text{mW}$ | $\chi^{(3)}$ $\text{esu}$ |
|--------------------|-------|---------------------------|-------|--|------------------------|-----------------|-------------------------------|-------|-------------------------------|---------------------------|
| $1 \times 10^{-5}$ | 57.96 | 151.6                     | 1.576 | 2.771                                      | 0.1                    | 0.0509          | $3.8 \times 10^{-11}$         | 0.605 | $2.8 \times 10^{-4}$          | $7.4 \times 10^{-5}$      |
| $2 \times 10^{-5}$ | 39.42 | 258.8                     | 1.964 | 2.340                                      | 0.11                   | 0.056           | $5.6 \times 10^{-11}$         | 0.455 | $4.7 \times 10^{-4}$          | $1.9 \times 10^{-4}$      |
| $5 \times 10^{-5}$ | 31.31 | 322.9                     | 2.09  | 2.127                                      | 0.17                   | 0.0866          | $8.5 \times 10^{-11}$         | 0.3   | $6.6 \times 10^{-4}$          | $3.1 \times 10^{-4}$      |
| $7 \times 10^{-5}$ | 27.21 | 361.9                     | 2.12  | 2.011                                      | 0.2                    | 0.1019          | $1.6 \times 10^{-10}$         | 0.126 | $8.7 \times 10^{-4}$          | $4.2 \times 10^{-4}$      |
| $1 \times 10^{-4}$ | 23.19 | 406.4                     | 2.63  | 1.889                                      | 0.21                   | 0.107           | $1.2 \times 10^{-10}$         | 0.1   | $9.6 \times 10^{-4}$          | $7.1 \times 10^{-4}$      |



#### 4. CONCLUSIONS

In conclusion, the nonlinear optical properties (refractive index, absorption coefficient, nonlinear susceptibility) for dye solution and solid sample (dye doped polymeric hosts) such as poly methyl methacrylate (PMMA) have been investigated by employing the EZ-scan technique. The experimental results show that the nonlinear refractive index for dye solution is positive while for thin film is negative. The nonlinear absorption coefficient for dye solutions behave like (Two photon absorption) while behave like (Saturation absorption) for thin film, also the nonlinear optical properties for solid samples are more clearer than in liquid samples, where polymeric samples be easier in measurement and more stabilizing than the liquid sample.

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