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Nonlinear Refraction and Optical Limiting Behavior of Eriochrome Cyanine R Dye Chromophore Under CW He-Cd Laser Illumination

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Abstract: ***The nonlinear optical refraction and optical limiting behavior of an organic dye, Eriochrome Cyanine R in aqueous solution, were investigated under excitation with cw He-Cd laser light at 442nm. The nonlinear optical responses of the material were studied, using single-beam Z-scan technique. Refractive nonlinearity of the samples was investigated using closed aperture Z-scan technique. All the samples exhibited a negative (defocusing) nonlinearity and large nonlinear refractive index of the order of 10^{-8} cm^2/W. The optical limiting behavior of the dye chromophore is also demonstrated.***

Keywords: Eriochrome Cyanine R; Nonlinear refraction; Z-scan;Optical Limiting;defocusing

1. Introduction

The rapid technological developments that took place in Optics during the last few decades have led to the search for materials with large optical nonlinearities, which would also satisfy all the technological requirements such as wide active optical frequency range, fast optical response time , high damage threshold, adaptability and capability to interfacing with other materials etc. Nonlinear opical (NLO) materials have great impact on information technology, industry, life sciences, and invariably in defence technology. Most of the current investigation made in nonlinear optical materials is motivated by its potential for high performance applications which are not accessible by electronics. Tremendous advance have been made in this field in a short period , driven by the current need for faster and higher means of communications and data processing and its potential in realizing all optical systems. One of the important applications of these materials is in optical limiting (OL) devices used to protect eyes and sensors against damage by exposure to sudden high-intensity light, which still remains a challenging problem. Optical limiters can transmit low-intensity laser pulses effectively and attenuate high-intensity laser pulses strongly by using the NLO properties of the materials [1].The selection of efficient materials is still the key point for optical power limiters and it has led to the study of materials that exhibit strong nonlinear absorption [2,3].

NLO organic molecules possess a strong donar-acceptor intermolecular interaction due to the presence of easily polarizable delocalized π -electrons in the system. Refractive nonlinearities associated with self-focusing and self-defocusing in various media are currently being explored with greater interest owing to their increasing application potential in various optical devices, including optical power limiters [4].

The organic dye Eriochrome Cyanine R (ECR; LobaChemie, Mumbai ,India) , belonging to triaryl methane family, is an important histological and bacterial stain and used for colouring textiles and leather. In this paper , we present the results of our investigations on the nonlinear refraction and OL response of the Organic chromophore, ECR (molecular structure shown in Fig.1) in aqueous solution under irradiation with 442 nm cw He-Cd laser light for three different dopant concentrations. The OL behavior was also demonstrated. Z-scan experiments and nonlinear transmission measurements were carried out to investigate the NLO properties and OL behavior, of the dye chromophore. The ECR solution samples displayed a negative refractive nonlinearity due to self-defocusing effect.

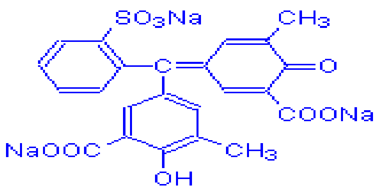


Figure 1: Molecular structure of Eriochrome Cyanine R dye molecule

2. Experimental

The fundamental optical constants required for the analysis of the z-scan data of the samples were obtained from the optical absorption measurements recorded using a ultraviolet-visible (UV-Vis) spectrophotometer (Shimadzu UV-2450). The nonlinear refraction behavior of ECR dye were investigated using single-beam z-scan technique. Z-scan method is a simple and effective tool for accurate determination of nonlinear refraction and its sign. The standard Z-scan set up [5], shown in Fig.2, was used for the measurement of NLO coefficient. The transmittance of the incident laser beam through an aperture in the far field was measured as a function of the sample position with respect to the focal plane. This formed a closed aperture scan. The experiment was performed at fixed input laser energy. The closed aperture scan is sensitive to nonlinear absorption as well as refraction[6]. OL experiments were also performed with ECR solution. The samples was kept at the focus of a converging lens (focal length 20 cm) and the output was collected through a lens kept behind an aperture having 90% transmittance.

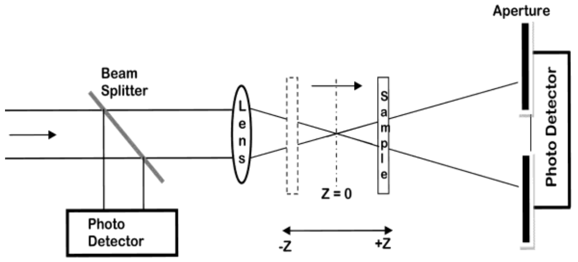


Figure 2: Schematic diagram of Z-scan experimental arrangement

3. Results and Discussion

The linear absorption spectra of ECR dye in aqueous solution for various dye concentrations are shown in Fig:3. The standard value of absorption maxima for ECR dye is 518nm. The observed values of absorption maxima for the samples for different concentrations are 517 nm. This shift in peak absorption wavelength is accounted for the structural and environmental changes occurring in the dye in polar solutions. The third-order nonlinear refractive index n_2, of the ECR dye in aqueous solution at various concentrations for the incident intensity I_0= (4.43 and 3.25) KW/cm^2 were evaluated by the measurements of Z-scan. The characteristic dependences of the normalized transmittance(T) of the sample as a function of sample position with respect to the focal point is shown in Fig:4(a-c) and Fig 5(a-c).The post-focal peak followed by a pre-focal valley normalized transmittance curve obtained from the closed aperture z-scan data, indicates that the sign of the refraction nonlinearity is negative,ie; self defocusing. The self defocusing effect is due to the local variation of the refractive index with temperature. The measurable quantity ΔT_{p-v} can be defined as the difference between the normalized peak and valley transmittances. The variation of this quantity as a function of $|\Delta\varphi_0|$ is given by

$$\Delta T_{p\_v} = 0.406(1-S)^{.25}|\Delta\varphi_0|$$

And $\Delta\varphi_0 = kn_2 I_0 L_{Eff}$

where$k = \frac{2\pi}{\lambda}$ is the wave vector and$\lambda$is the wave length of the laser, all in free space.

$$S = 1 - \exp(-\frac{2r_a^2}{w_a^2})$$

Here S is the linear transmission of the aperture, $r_a$is the aperture radius and w_a, the beam radius at the aperture in the linear regime [7]. Here $\Delta\varphi_0$ is the on-axis phase shift at the focus, I is the radiation intensity, L_{eff} is the effective length of the sample which is equal to $L_{eff} = (1 - e^{-\propto L})/\propto$ With L sample length and in $\propto$ linear absorption coefficient.

Experimentally determined nonlinear refractive index n_2 can be used to find the real part of the third-order nonlinear optical susceptibility [χ^3] according to the following relation[8],

$$Re\ \chi^3 = \frac{10^{-4}\epsilon_0(n_0{}^2)c^2 n_2}{\pi} \text{ (cm}^2\text{/W)}$$

Where ϵ_0 is the vaccum permitivity and C is the light velocity in vacuum.

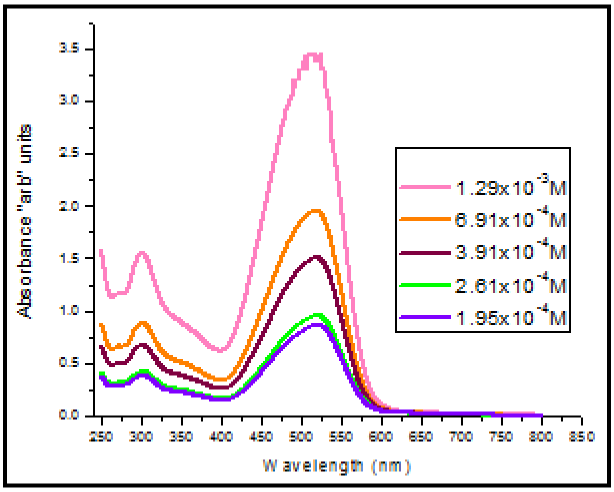
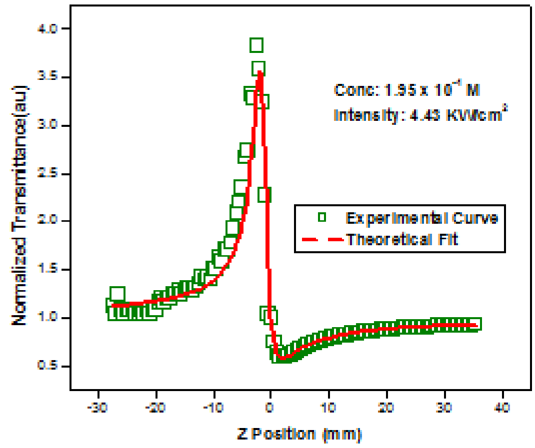


Figure 3: UV- Visible Absorption Spectra of Eriochrome Cyanine R in Solution

The experimentally determined values of ΔT_{p-v},n_2 ,$Re\ \chi^3$ are given in Table 1.

Fig: 6 shows the optical limiting behavior of the dye, in the power range 10.23-57.6 mW. The straight line in the figure represents the linear transmission(67.3%). It can be seen that at very low incident laser intensities, the transmission linearly increases, obeying Beer's law, and at high incident intensities the transmission starts to deviate from linearity. A low limiting threshold is highly desirable for protection of human eyes; for , eye's retina can be permanently damaged on exposure to laser beam of even a few mW from a laser pointer for a duration as short as .25s [9,10].



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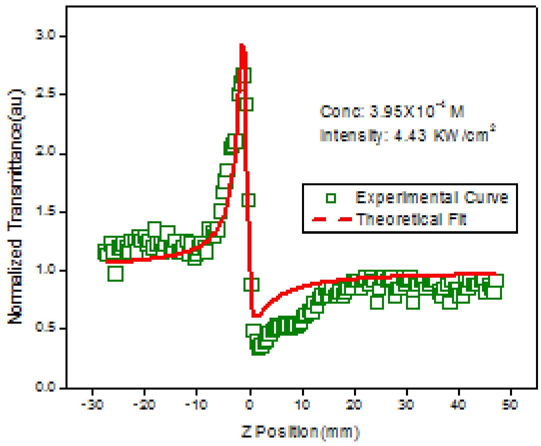

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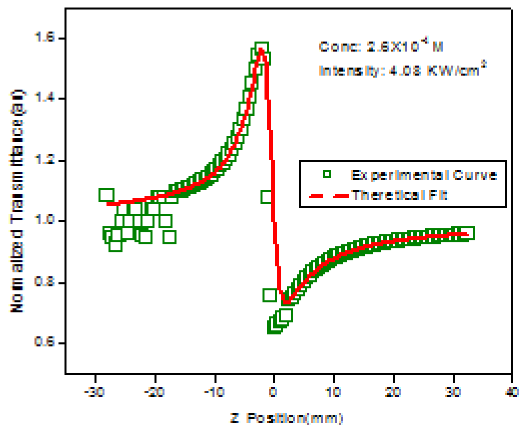

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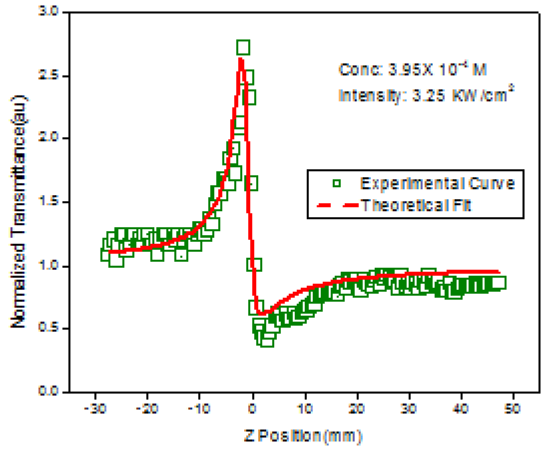
Figure 4(a-c):- CA Z-Scan profiles of EC R in aqueous solution with Concentrations $1.95X10^{-4}$ M , $3.95X10^{-4}$ M and $2.61X10^{-4}$Mat I_0= 4.43 KW/cm^2 .



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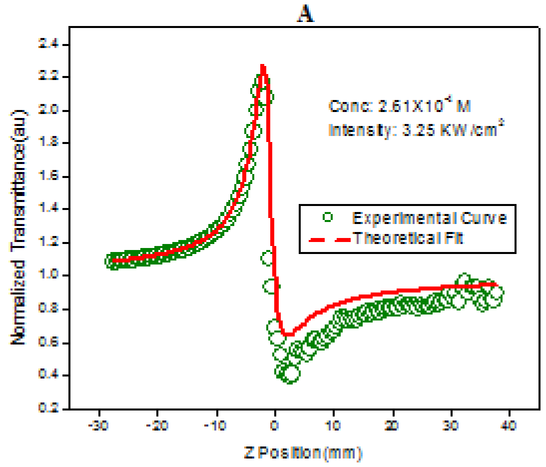

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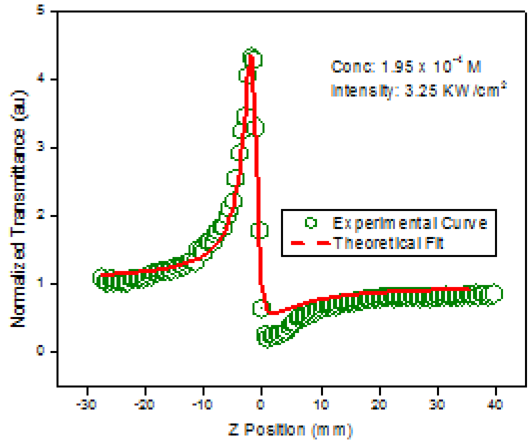

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Figure 5(a-c): CA Z-Scan profiles of EC R in aqueous solution with Concentrations $1.95X10^{-4}$ M , $3.95X10^{-4}$ M and $2.61X10^{-4}$M at I_0= 3.25 KW/cm^2

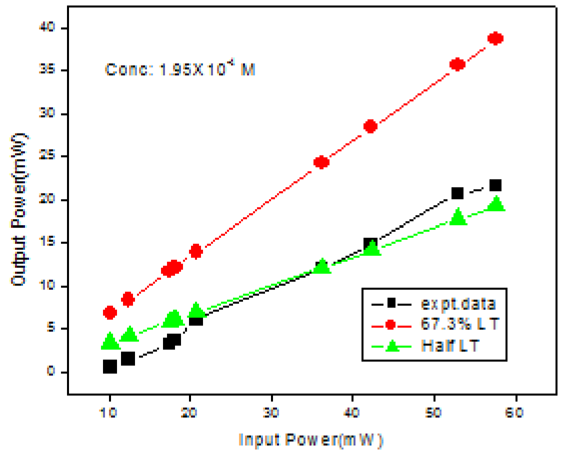

Figure 6: Optical Limiting behavior of EC R in aqueous solution having concentration $1.95x10^{-4}$ M.

Table 1: The values of n_2 and $Re\ \chi^3$ for three different concentrations of ECR dye in aqueous solution

Concentration ($\times10^{-4}$ M)	I_0 (KW/cm^2)	ΔT_{p_v}	n_2($\times 10^{-8}$ cm^2/W)	$Re\ \chi^3$ $X10^{-6}$ (esu)
1.95	4.43	1.522	3.272	1.47
	3.25	1.368	4.0092	1.804
2.61	4.43	2.017	4.5406	2.041
	3.25	2.011	5.27	2.37
3.95	4.43	2.71	6.36	2.86
	3.25	2.32	7.31	3.32

4. Conclusion

In summary, aqueous solutions of the organic dye EC R, were prepared for three different concentrations and its low-threshold NLO properties were investigated. The effect of dye concentration on the absorption spectra was studied using UV-Visible spectrophotometer and found to be in well agreement with the Beer-Lambert law.

The third order nonlinear refraction behavior was studied using the single beam Z-scan technique, employing the 442 nm cw He-Cd laser as excitation source. The dye samples exhibited a negative refractive nonlinearity due to the self-defocusing effect. The nonlinear coefficient of the refractive index (n_2) at this wavelength was also evaluated. The

Optical limiting response of the dye samples was also demonstrated.

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