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Nonlinear optical properties and diffraction ring patterns of benzo congo red

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ABSTRACT

The nonlinear optical properties of Benzo congo red in Dimathyl sulfoxide (DMSO) solvent and polymer were studied using single beam Z-scan technique with a continuous-wave radiation at 532 nm of an output power of 40 mW. All the solution samples and polymer showed large nonlinear refractive indexice and absorption coefficient of the order of 10^{-8} cm^2/W and 10^{-3} cm/W, respectively. The concentration-dependent nonlinear refractive index was also investigated. We presents experimental evidences of observing diffraction pattern in congo red dye: DMSO solvent with the calculation of the refractive index change, Δn , the relative phase shift, $\Delta\Phi$, and effective nonlinear refractive index ,n_2. The solvent of spectroscopic grade and was used without further purification. All the solutions used for the study were freshly prepared.

Keywords*:* nonlinear refraction index , Z-scan, azo dye, cw laser.

INTRODUCTION

Azo dyes have many advantages over other nonlinear optics (NO) materials. Photoisomerization of azo molecules enables modifies their linear and nonlinear polarizability of them as well as optical nonlinear refraction. Since the optical properties of azo molecules can be controlled optically, it has intrigued considerable interest of people [1,2]. The nonlinear optical phenomena of azo dyes can result from electronic response and/or nonelectronic one. The electronic nonlinearity is induced by either population redistribution or distortion of electronic clouds. A molecule undergoes a transition from its ground state to its excitation state after absorbing a photon. The dipole moment of the molecule changes during such a transition. The change in the dipole moment will give birth to electronic nonlinearity. A nonelectronic response is a non-radiative interaction such as cis-trans isomerism, the changes in density and temperature [3–5]. It has been well known that the nonlinear optical behavior of materials can vary greatly by changing different laser duration or different laser wavelengths. Thus, studies about the mechanism of their nonlinear optical response with different laser duration or different laser wavelengths are expected to be more interesting and important. If the nonlinear mechanism is understood for certain laser pulses, the NLO properties optimization can be well accomplished. Z-scan technique is a simple and effective tool to determine the nonlinear properties [6]. It has been widely used in material characterization because it provides not only the magnitudes of the real part and imaginary part of the nonlinear susceptibility, but also the sign of the real part. Both nonlinear refraction and nonlinear absorption in solid and liquid samples can be measured easily by Z-scan technique, which use the change of transmittance of nonlinear materials [5].

In this work, we demonstrate the optical nonlinearities of a Benzo congo red at different concentration in Dimathyl sulfoxide (DMSO) and thin film through Z-scan technique under laser excitation at 532 nm cw solid state laser with an output power of 40 mW and presents experimental evidences of observing diffraction pattern in Benzo congo red:

DMSO solvent with the calculation of the refractive index change, Δn , the relative phase shift, $\Delta\Phi$, and effective nonlinear refractive index ,n_2 .

MATERIALS AND METHODS

Synthesis of dye doped polymer film

Congo red, from azo dye family was chosen for the study. The molecular structure of the dye is shown in Fig. 1. The dye-doped PMMA films were prepared as follows: dye and PMMA are dissolved separately in Dimathyl sulfoxide and then the solution of dye and that of PMMA are mixed, the mixture was stirred at room temperature (RT) for 30 min. to inter all Benzo congo dye molecules within polymer chains, then the solution were filtered through 0.2µm syringe filter. After that the solution of Congo red: PMMA were mixed, heated (up to 65 ^{o}C) and stirred for (50 min.), the film were prepared on a clean glass slide by the repeat-spin-coating method and dried at room temperature (300K) for 48 h. The optical quality of the film is checked by passing He-Ne laser beam of power of 5mW. Film which shows no distortion or dispersion of the laser beam alone is taken for further studies. The DDP film of concentration 0.07 mM and thickness of 0.15 µm was synthesized.

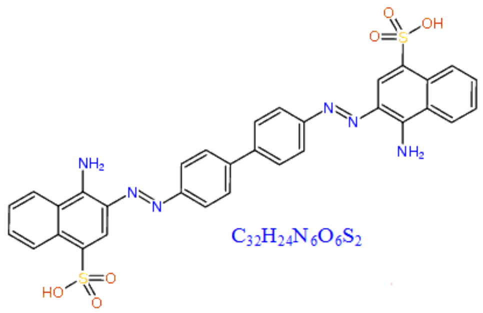

Fig.1: Chemical structure and molecular formula of Benzo congo red dye.

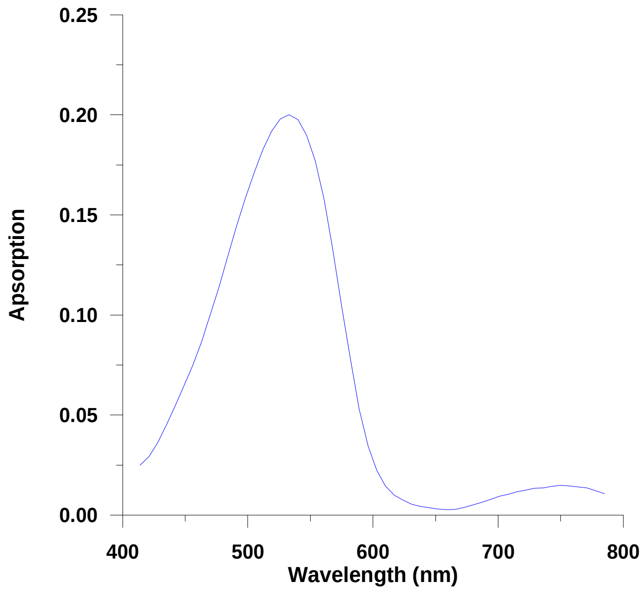

Fig. 2: UV-VIS absorption spectrum Benzo congo red dye in DMSO solvent.

Absorption spectra

The solution samples of the azo dye were prepared in DMSO. The former was contained in a 1mm quartz cuvette. The linear absorption spectrum of the azo dye solution with the concentration of 0.06 mM in DMSO solvent is shown in Fig.2, which was acquired using a UV–VISNIR spectrophotometer (Type: CECIL –CE-3550) .

The Z-scan experiments were performed using a 532 nm solid state laser beam, which was focused by 50 mm focal length lens. The laser beam waist ω_0 at the focus is measured to be 19.36 µm and the Rayleigh length z_R =4.06 mm. The schematic of the experimental set up used is shown in Fig. 3. A 1mm wide optical cell containing the solution of Benzo congo red dye is translated across the focal region along the axial direction that is the direction of the propagation laser beam. The transmission of the beam through an aperture placed in the far field was measured using a photodetector fed to the digital power meter. For an open aperture Z-scan, a lens was used to collect the entire laser beam transmitted through the sample replaced the aperture.

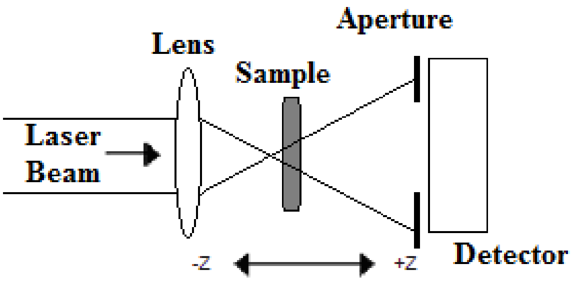

Fig. 3. Schematic diagram of experimental arrangement for the Z-scan measurement

RESULTS AND DISCUSSION

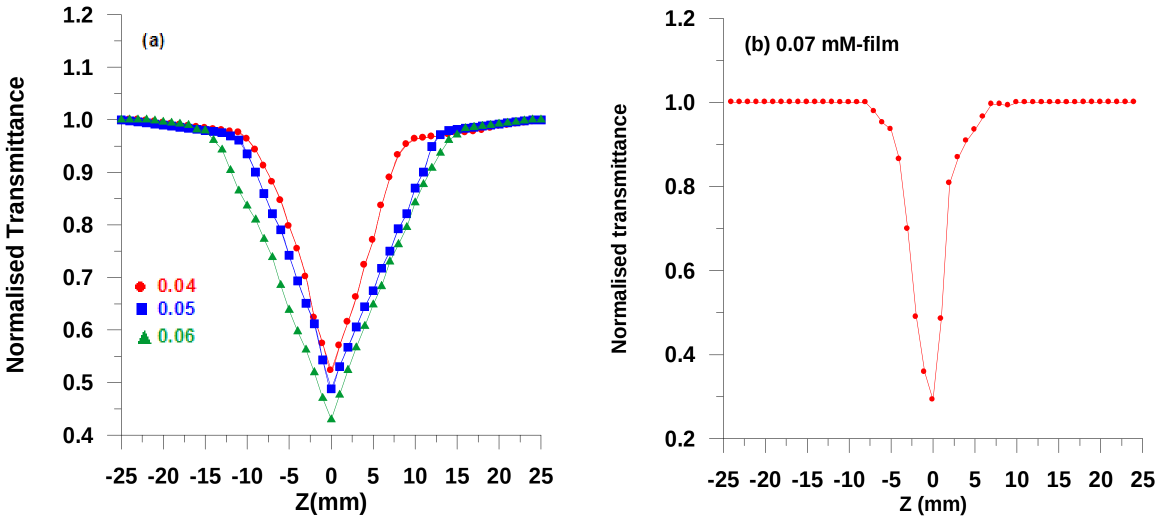

Fig. 4(a): Open-aperture Z-scan data (a) azo dye solutions at different concentration (b) azo dye doped polymer.

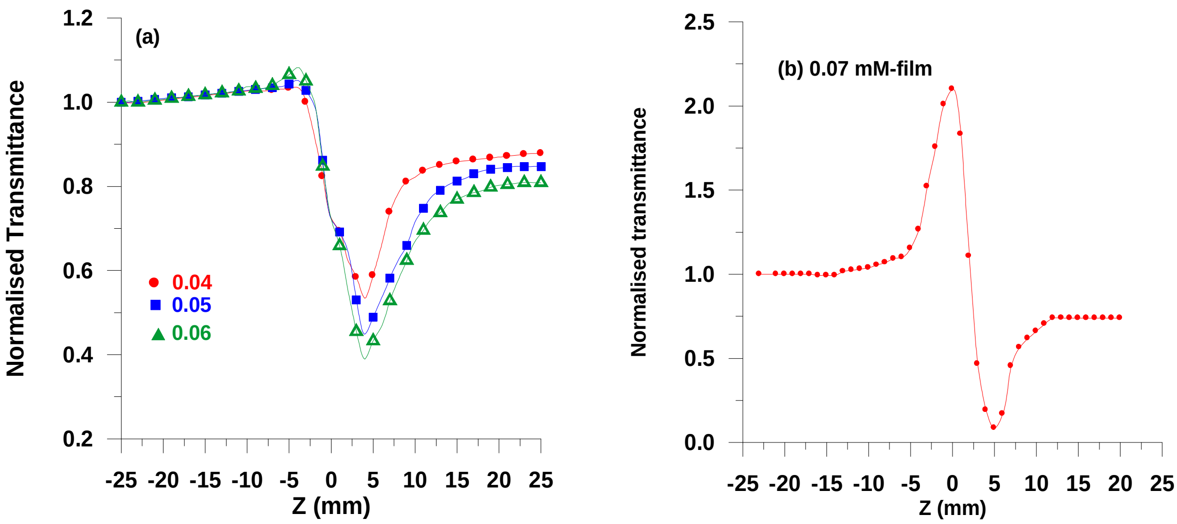

Fig. 5(a): Closed-aperture Z-scan data (a) azo dye solutions at different concentration (b) azo dye doped polymer.

1. 1. Z-scan Measurements:

The third-order nonlinear refractive index n_2 and the nonlinear absorption coefficient β, of the azo dye in DMSO at various concentrations for the incident intensity I_0 = 6.792 kW/cm^2 were evaluated by the measurements of Z-scan. Figure 4a and b shows the open Z-scan curve for the dye in solution at various concentrations and dye doped polymer film. The typical Z-scan data with fully open aperture is insensitive to nonlinear refraction; therefore, the data is expected to be symmetric with respect to the focus, but absorption saturation in the sample enhances the peak and decreases the valley in the closed aperture Z-scan curve and results in distortions in the symmetry of the Z-scan curve about Z = 0 [7].

The measurable quantity ΔT_{P-V} can be defined as the difference between the normalized peak and valley transmittances, $T_P - T_V$. The variation of this quantity as a function of $|\Delta\Phi_0|$ is given by [8]

$$\Delta T_{P-V} = 0.406(1-S)^{0.25}|\Delta\Phi_0|$$

Z-scan with a fully open aperture (S=1) is insensitive to nonlinear refraction (thin sample approximation). The aperture linear transmittance is given by $S = 1 - \exp(-2r_a / \omega_a)$, with r_a =2.5 mm the aperture radius ,ω_a = 5 mm the radius of the laser spot before the aperture, $\Delta\Phi_0$ is the on-axis phase shift. The on axis phase shift is related to the third-order nonlinear refractive index by

$$\Delta\Phi_0 = kn_2 I_0 L_{eff}$$

where $k = 2\pi / \lambda$ is the wave number and $L_{eff} = (1-\exp(-\alpha_0 L))/\alpha_0$ is the effective thickness of the sample , α is the linear absorption coefficient, L the thickness of the sample, I_0 the on-axis irradiance at focus and n_2 the third-order nonlinear refractive index. The defocusing effect of the dye in solution at various concentrations are shown in Fig. 5a and b. This defocusing is attributed to a thermal nonlinearity resulting from absorption of radiation at 532 nm. Localized absorption of a tightly focused beam propagating through an absorbing dye medium produces a spatial distribution of temperature in the dye solution and, consequently, a spatial variation of the refractive index, that acts as a thermal lens resulting in severe phase distortion of the propagating beam.

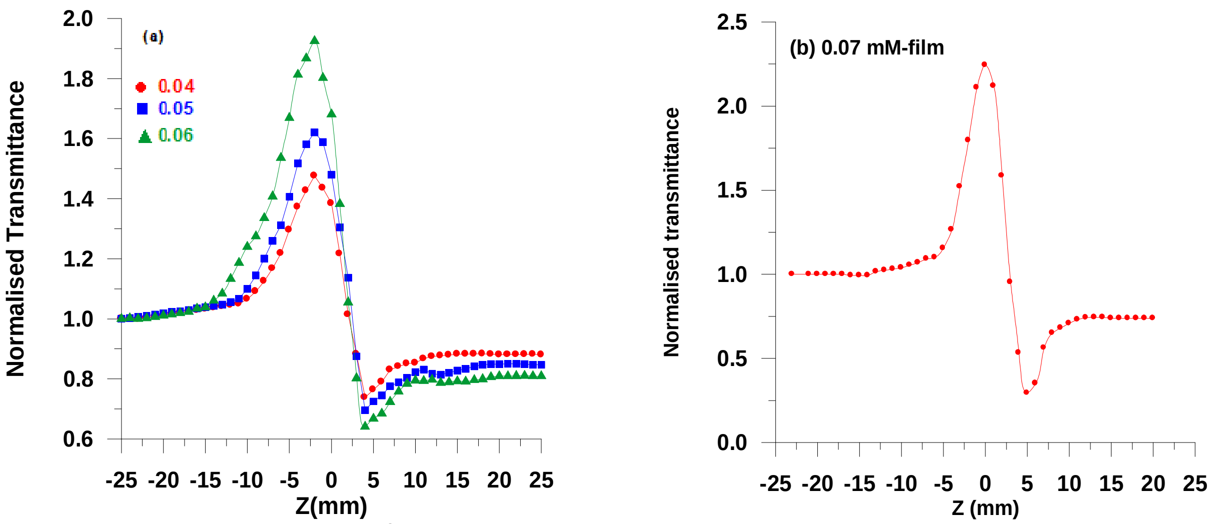

Fig. 6(a): Pure nonlinear refraction curve dye in solvent at various concentration (b) dye doped polymer.

Generally the measurements of the normalised transmittance versus sample position, for the cases of closed and open aperture, allow determination of the nonlinear refractive index , n_2 , and the reversible saturation absorption

(RSA) nonlinear coefficient, β, [9,10]. Here, since the closed aperture transmittance is affected by the nonlinear refraction and absorption, the determination of n_2 is less straightforward from the closed aperture scans. Therefore, it is necessary to separate the effect of nonlinear refraction from that of the nonlinear absorption. A simple and approximate method [11] to obtain purely effective n_2 is to divide the closed aperture transmittance by the corresponding open aperture scans.

With an open aperture the sample's transmittance is related to the nonlinear absorption coefficient through the relation [12]:

$$T(z) = 1 - \frac{q_0}{2\sqrt{2}} \qquad \text{For } q_0 < 1$$

where

$$q_\circ(z) = \frac{\beta I_\circ (1 - e^{-\alpha L})}{(1 + z^2 / z_o^2)\alpha}$$

in which β is the nonlinear absorption coefficient, α is the linear absorption coefficient, L is the thickness of the sample, I_0 is the intensity of the laser beam at the focus (z = 0), and z_0 is the Rayleigh radius. In the Gaussian approximation, z_0 is related to the beam waist through the relation $z_0 = \pi\omega_0^2/\lambda$, where λ is the wavelength. The experiment was repeated for the pure solvent Dimathyl sulfoxide to account for its contribution, but no significant measurable signals were produced in either the open or the closed Z-scan traces. The nonlinear parameters calculated are as tabulated in Table 1.

Table.1. Nonlinear parameters of Benzo congo red dye solutions and polymer

Concentration(mM)	**ΔΦ**	$\Delta n x10^{-4}$	$n_2 x10^{-8} cm^2/W$	$\beta x10^{-3} cm/W$
0.04- Liquid	**1.86**	**2.17**	**3.19**	**2.29**
0.05- Liquid	**2.33**	**2.90**	**4.27**	**2.62**
0.06- Liquid	**3.24**	**4.11**	**6.05**	**2.97**
Polymer film 0.07	**5.16**	**6.47**	**9.53**	**4.55**

2. Diffraction Ring Patterns Measurements

The experimental setup for the diffraction ring patterns are the same as mentioned, expect that's the power meter detector is replaced by the transparent screen. We can estimate the induced refractive index change, Δ*n*, and the effective nonlinear refractive index, n_2, for the preceding data as follows. Because the laser beam used in the experiment has a Gaussian distribution, the relative phase shift, $\Delta\Phi$, suffered by the beam while traversing the sample of thickness (L) can be written as [13]:

$$\Delta\Phi = kL\Delta n \qquad (1)$$

Where *k*=2π/λ is the wave vector in vacuum and λ is the laser beam wavelength.
The on-axis nonlinear phase-shift, $\Delta\Phi$, can be related to the number of rings , N, observed as [14]

$$\Delta\Phi = 2\pi N \quad \text{...(2)}$$

Δn can be related too to the total refractive index of the medium , n, and the back ground refractive index , $n_\circ$, as [12]

$$n = n_\circ + \Delta n \quad \text{...(3)}$$

where n_0 is the background refractive index.
and

$$\Delta n = n_2 I \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (4)$$

By the combination of equations (1-4) one can calculate, Δn , $\Delta\Phi$ and n_2 .
As given in Table 2,the number of rings N for 100 mW power, observed are 14,16 and 19 respectively while for 90 mW power the number of rings N are 7,13 and 15 respectively. The diffraction ring patterns for the azo dye solutions are shown in Fig.5

Table2: Nonlinear parameters of Benzo congo red dye solutions using diffraction ring patterns

Con.(mM)	Rings No.at 100mW	ΔΦ	n_2 (cm^2/W)	Δn	Rings No. at 90 mW	ΔΦ	n_2 (cm^2/W)	Δn
0.04- Liquid	14	1.53	1.10×10^{-8}	1.49×10^{-4}	7	0.76	0.61×10^{-8}	0.74×10^{-4}
0.05- Liquid	16	1.75	1.34×10^{-8}	1.82×10^{-4}	13	1.42	1.21×10^{-8}	1.48×10^{-4}
0.06- Liquid	19	2.08	1.62×10^{-8}	2.21×10^{-4}	15	1.64	1.42×10^{-8}	1.74×10^{-4}

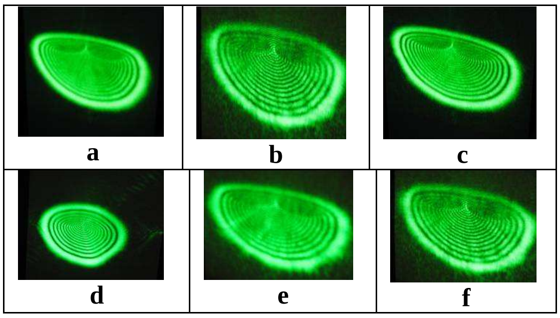

Fig.5 diffraction ring patterns for the Benzo congo red solutions (a)14, (b) 16,(c) 19 for 100mW and (d) 7, (e) 13 ,(f) 15 for 90mW.

CONCLUSION

In summary, we have measured the nonlinear refraction index coefficient n_2 and the nonlinear absorption coefficient β for solutions of Benzo congo red dye for various concentrations and polymer using the Z-scan technique with 532 nm of solid state laser (SDL). The Z-scan measurements indicated that the dye exhibited large nonlinear optical properties. We have shown that the nonlinear absorption can be attributed to a saturation absorption process, while the nonlinear refraction leads to self-defocusing in this dye. All the solutions samples showed a large nonlinear refractive index of the order of 10^{-8} cm^2/W and 10^{-3} cm/W, respectively. Experimental results of ring patterns suggest the possibility of using Benzo congo red dye for various concentrations solvent in DMSO in all optical systems .These patterns were generated in Benzo congo red solution by the irradiation with visible laser beam of Gaussian extent .The instantaneous formation of rings prove the fast response of this substance .The thermal number of rings observed increases with increasing input power nonlinearly .The stability of the ring patterns suggest the stability of such medium. The calculation of the refractive index change, Δn , the relative phase shift, $\Delta\Phi$, and effective nonlinear refractive index ,n_2 were measured too.

All these experimental results show that the solution of Benzo congo red dye and Benzo congo red doped polymer thin film are a promising material for applications in nonlinear optical devices.

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