Advances in Physics Theories and Applications
ISSN 2224-719X (Paper) ISSN 2225-0638 (Online)
Vol.26, 2013

Z-Scan Measurement For The Thermo-Optic Coefficient and Transmitted Beam Profile Of 1.8-Dihydroxy-Naphthalin-3, 6 Disulfonic Acid-[2-(4-azo)]-N-(5-Methyl-3-Isoxazolyl)-Benzene Sulfonamide

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The research is financed by Asian Development Bank. No. 2006-A171(Sponsoring information)

Abstract

The nonlinear optical properties of an azo dye (1.8-Dihydroxy-naphthalin-3, 6 disulfonic acid- [2- (4-azo)]-N-(5-methyl-3-isoxazolyl)-benzene sulfonamide) are studied by using z-scan and diffraction ring technique with continuous wave (cw) laser at a wavelength of 532 nm. The obtained results for the nonlinear refractive index , n_2, and the thermo-optic coefficients , dn/dT, are found to be of the order of 10^{-8} Wcm^{-2} and 10^{-6} K^{-1} respectively. The transmitted beam profiles, the distribution of intensity corresponding to the sample positions and D- distribution of rings number of each pattern variation for the azo dye samples have been studied. These results indicate that the azo dye is a promising candidate for applications in nonlinear optical devices.
Keywords: Nonlinear refractive index, Thermo-optic coefficients, Diffraction ring.

1. Introduction

In recent years, extensive studies have been carried out on organic nonlinear optical (NLO) materials due their very high nonlinearity, less dense, chemical stability and short response time to optical excitation properties irrespective of their poor mechanical and thermal properties (Gandhimathi & Dhanasekaran 2012). NLO materials can be used to manipulate optical signals in telecommunication systems and other optical signal processing applications (Ogawa *et al* 2002). Organic materials are considered as one of the important classes of third-order NLO materials because they exhibit large and fast nonlinearities and are, in general, easy to process and integrated into optical devices (Bredas *et al*. 1995; Rik *et al*. 1998; Gubler 2002). Moreover, a fine-tuning of the NLO properties of organic compounds can be achieved by rational modification of the chemical structure (Gema *et al*. 2004). Various types of organic compounds have been studied to obtain materials with large third-order nonlinearity. On the other hand a wide range of techniques have been used to measure third-order nonlinearity: e.g. Z-scan (Sheik-Bahae *et al*.1989 ; Sheik-Bahae *et al*.1990), nonlinear interferometry (Moran *et al*. 1975), degenerate four-wave mixing (Qussaiy *et al*. 2006), nearly degenerate three-wave mixing (Adair et al.1987), ellipse rotation (Owyoung 1973), beam distortion measurements (Williams *et al*. 1984), optical third harmonic generation (THG) (Maker & Terhune 1965) and frequency resolved optical gating (Wang *et al*.1999).
Materials that possess nonlinear optical properties have been investigated extensively for their potential applications in optical fibers, data storage, optical computing, optical switching, and optical limiting (Lanzerotii *et al*.1996; Lidorikis et *al*.1997; Justus *et al*.1993; Alan *et al*.1993). Among the promising class of materials, azo dyes (Katz *et al*.1987; Brzozowski *et al*. 2001; Rangel-Rojo *et al*.1998; Yang *et al*. 2005) play a vital role because of their good photo-thermal stability, dissolvability etc. The character of its molecular structure is double-bond N= N between the two phenyls (Shengwen *et al*. 2004; Yildiz *et al*. 2002; Mendez *et al*. 2005). Its potential application is to work as novel optical limiter for its nonlinear optics effect. The extensive use of continuous wave lasers for various applications with power levels ranging from μW to kW has induced a need to protect the human eyes and sensors (Gayathri & Ramalingam 2008). In order to find the suitability of a material for nonlinear applications one needs to study its photo physical as well as its optical characteristics such as type of nonlinearity, its magnitude, response time etc . In this article, For azo dye (1.8-Dihydroxy-naphthalin-3, 6 disulfonic acid-[2-(4-Azo)]-N-(5-methyl-3-isoxazolyl)-benzene sulfonamide) portability in the form complexes with some metals, we have chosen vanadyle ion (VO^{+2}) for this study because of its importance, , existence in nature and its composition stable complexes in oxidative status (+4). The synthesis of the chosen azo dye is shown in Figure1. We report the results of the refractive nonlinearities studied by using the single beam Z-scan

technique on low-cost azo dye in Dimethyl sulfoxide (DMSO) solvent in their resonant region using a continuous wave 532 nm diode-pumped laser. We presents experimental evidences of observing diffraction pattern in pure azo dye and azo dye with vanadyle ions (VO^{+2}) with the calculations of the effective nonlinear refractive index ,n_2, and variation of refractive index with temperature ,dn/dT.

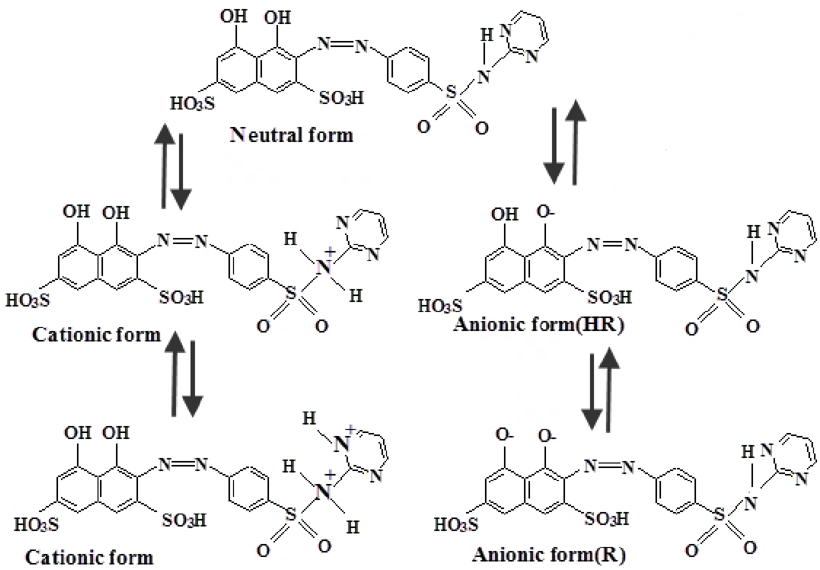

Figure 1. synthesis of the azo dye.

2. Experimental

2.1. UV–visible Spectroscopic studies

A UV–visible spectroscopy has been used to characterize the azo dye (1.8-Dihydroxy-naphthalin-3, 6 disulfonic acid-[2-(4-Azo)]-N-(5-methyl-3-isoxazolyl)-benzene sulfonamide) in the spectral range (300–700nm). The absorbance (A) of the sample measured using Cecil Reflected-Scan CE 3055 reflectance spectrometer. These measured was performed at room temperature. The optical absorption of the dye and dye with VO^{+2} in DMSO Solvent with 0.08 mM concentration shows an absorption peak (λ_{max}) at 520 and 527 nm, respectively as shown in Figure 2. Also we can see from the Figure 2 that the optical absorption for azo dye with VO^{+2} increases more than pure azo dye this due to increase number of molecular per unit volume, so the absorbance will be increased.

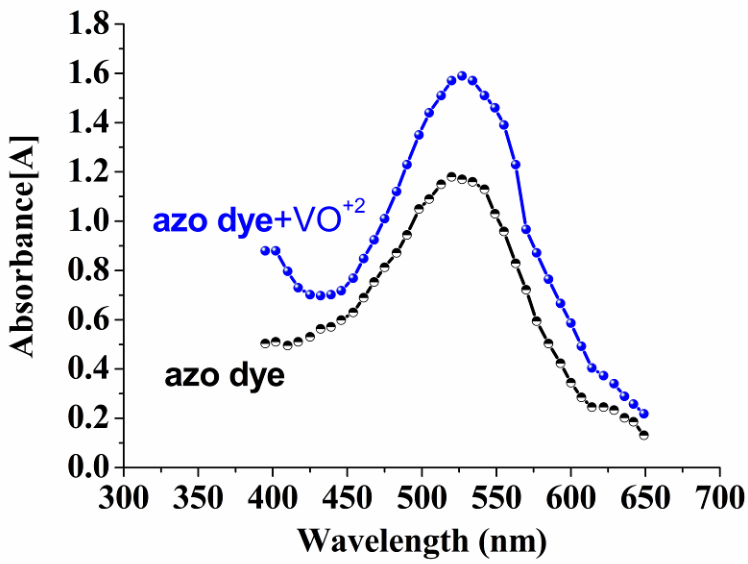

Figure 2. UV-VIS absorption spectrum of azo dye at 0.08 mM concentrations.

2.2. Z-scan measurement

The Z-scan technique was used to determine the nonlinear optical properties of the investigated sample. This technique is a simple and sensitive method for measurement of nonlinear refractive indices and nonlinear absorption of nonlinear optical materials. In this technique, the sample scans along the optical axis (designated the z direction) in the focal region of a single focused laser beam, and the transmission of the laser beam through an aperture placed in the far field was measured using a photo detector fed to a power meter. The experimental setup used is shown in Figure 3. A 1 mm quartz cell containing the dye in DMSO was translated across the focus of the lens along the direction of the propagation of laser beam.

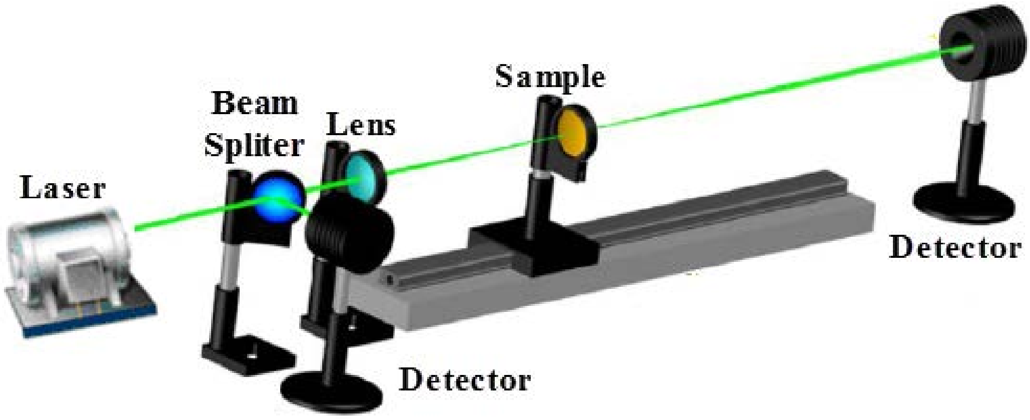

Figure 3. The optical geometry for measuring nonlinear optical responses.

A beam from continuous SDL laser operating at 532 nm and power of 18 mW is used to perform the measurement. The beam was focused on the sample using +5 cm focal length lens.

Figure 4 shows the closed aperture Z-scan data of pure azo dye in chloroform solvent and azo dye with VO^{+2} at 0.08 mM concentration at incident intensity I =2.449 KW/cm^2 .

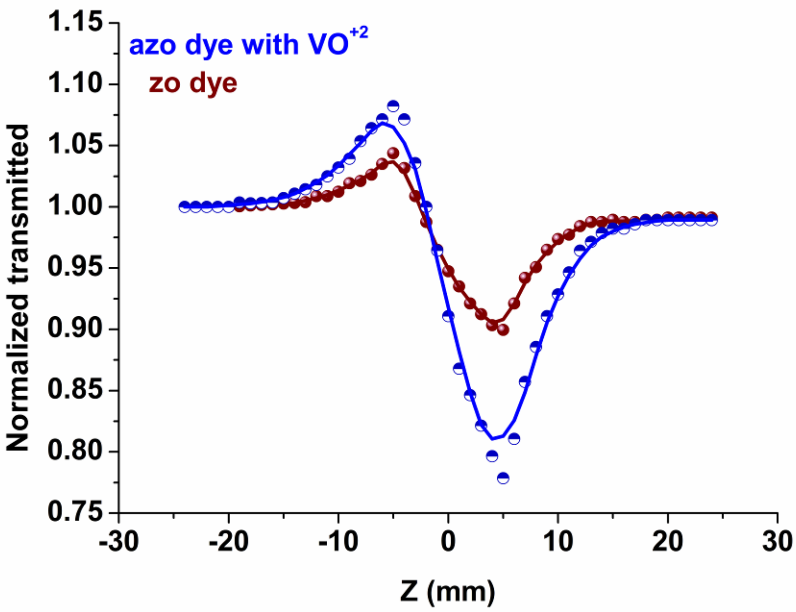

Figure 4 . Closed aperture Z-Scan data for azo dye solution.

The beam waist at the focal point was estimated to be 21.63 μm and the corresponding Rayleigh range was z_0 = 2.76 mm. The sample was moved along the z-axis using a translation stage. An aperture of 5 mm diameter was mounted in front of the photo detector placed about 10 cm away from the beam focus. The intensity transmitted by the sample was measured as a function of the sample position along the z-axis, there by obtained the closed aperture data. The measurements were repeated after removing the aperture in order to obtain the open aperture data. Figure 5 shows the measured Z-scan data for open aperture set-up for the pure azo dye in solvent and azo dye with VO^{+2} at 0.08 mM concentration.

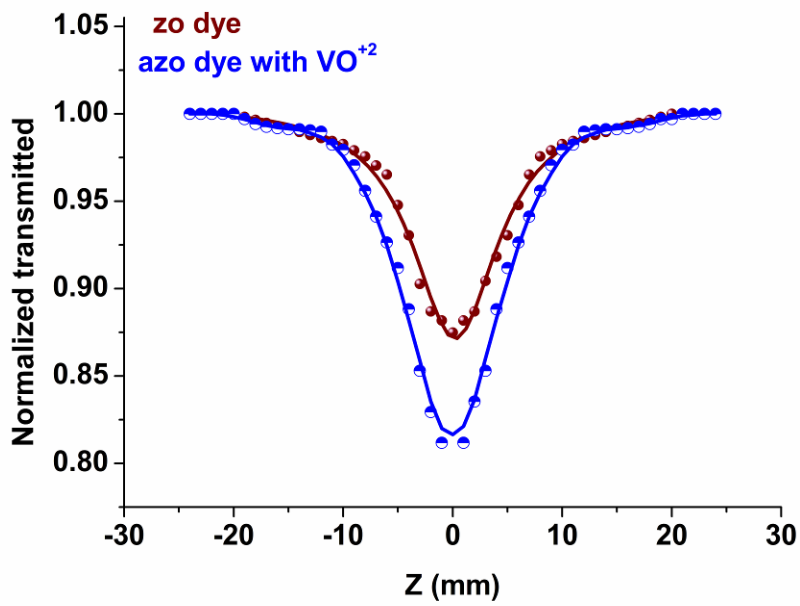

Figure 5. Open aperture Z-Scan data for azo dye solution.

For a purely refractive nonlinearity, the amplitude of the transmitted intensity changes as a function of the sample position. The nonlinear medium acts as a positive lens (for $n_2 > 0$) or a negative lens ($n_2 < 0$) (Fryad, 2011). Thus, a prefocal transmittance maximum (peak) which is followed by a post focal transmittance minimum (valley) is the z-scan signature of negative nonlinear refraction. Positive nonlinear refraction, by the same analogy, gives rise to an opposite valley–peak configuration. The simplest way to do this is varying the beam size by translating the sample through the focal point. The laser source is assumed to have a Gaussian beam profile so that the electric field variation is (Majles Ara *et al*. 2009).

$$E(z,r,t) = E_0(t)\frac{\omega_0}{\omega(z)}\exp\left(-\frac{r^2}{\omega^2(z)} - \frac{ikr^2}{2R(z)}\right)\exp(-i\varphi(z,t)). \qquad (1)$$

where $\omega^2(z) = \omega_0^2(z)(1+\frac{z^2}{z_0^2})$ is the spot size in z , $R(z) = (1+\frac{z_0^2}{z^2})$ is the radius curvature , and

z_0 is the diffraction length of the beam $z_0 = k\omega_0^2/2$.

The nonlinear refraction index, n_2, can be calculated from the peak-to-valley height ($\Delta T_{p\text{-}v}$) of the normalized smoothed curve of experimental points as follow (Badran 2012):

$$\Delta T_{P-V} = 0.406(1-S)^{0.25}\Delta\varphi_0 \qquad \Delta\varphi_0 < \pi \qquad (2)$$

where S is the linear transmission of the aperture and is given by (Rekha & Ramalingam 2009):

$$S = 1-\exp\left(-\frac{2r_a^2}{\omega_a^2}\right) \qquad (3)$$

where r_a = 2.5 mm is the radius of the aperture and ω_a =6.28 mm is beam radius at the aperture in the linear region.

Because the laser beam used in the experiment has a Gaussian distribution, the relative plane distortion, $\Delta\varphi_0$, suffered by the beam while traversing the sample of thickness, L_{eff} can be written as (Badran *et al*. 2012):

$$\Delta\varphi_0 = \frac{2\pi L_{eff} n_2 I_0}{\lambda} \tag{4}$$

where $k = 2\pi/\lambda$ is the wave vector in vacuum and λ is the laser beam wavelength , L_{eff} is the effective thickness of the sample $L_{eff} = (1-e^{-\alpha L})/\alpha$ and α is the linear absorption coefficient. The ratio of Figures. 4 and 5 scans is shown in Figure 6. The values of the linear absorption and nonlinear refractive index of azo dye and azo dye with VO^{+2} in DMSO solvent at 0.08 mM concentration are given in Table1.

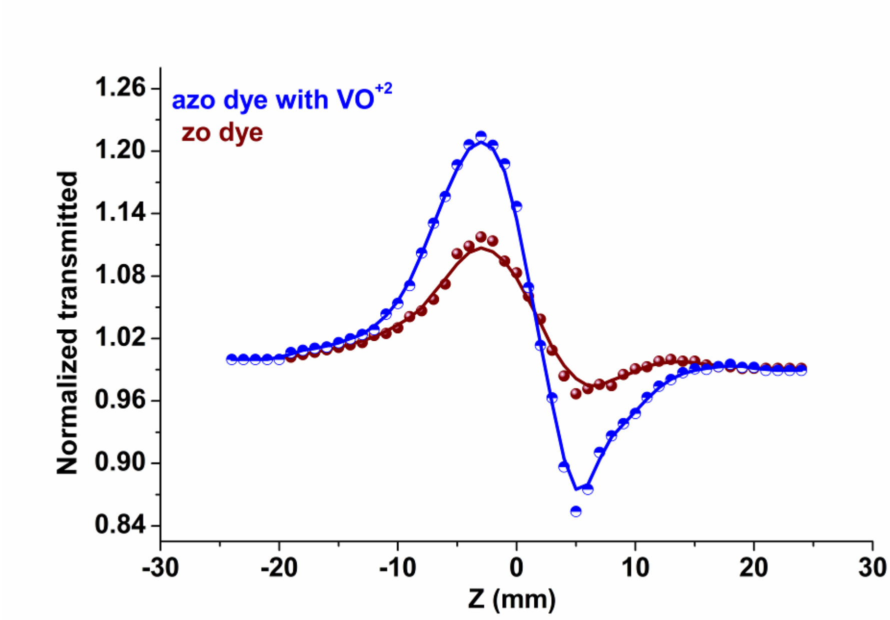

Figure 6. Pure nonlinear refraction curve for azo dye solution.

Table1. The linear absorption coefficient and nonlinear optical parameters.

Sample	$\alpha\ cm^{-1}$	$\Delta\varphi_0$	n_2 (cm^2/W) ×10^{-8}	dn/dT (k^{-1}) ×10^{-6}
azo dye	26.714	0.4018	3.988	1.999
azo dye +VO^{+2}	36.387	0.9607	12.417	4.569

In nonlinear optical phenomena the spatial self-phase modulation on the cross-section of the Gaussian beam emerges as a kind of wave front distortion. For a thin nonlinear medium, although the change in the radial size of the Gaussian beam caused by the self-focusing and defocusing is negligible, the spatial self-phase modulation induced by the self-action is rather appreciable. For a beam with a Gaussian profile the phase increment $\Delta\varphi_0$ has a bell-shaped distribution of which the centre is at *r* = 0. If $(\Delta\varphi_0)_{\max}$ is much larger than 2*π*, a set of concentric rings will appear on the far-field observation screen as the Gaussian beam is transmitted through the nonlinear medium (Deng *et al*. 2005). The spot of the transmitted beam was photographed at far away distance from the sample, when the sample was at different positions. Figure 7(a)–(d) shows the distribution of intensity for pure azo dye solution and Figure 8(a)–(d) shows the distribution of intensity for azo with Vo^{+2} solution, when the samples was far from the focus (Z = -10 mm), at the focus (Z = -4 mm),out of focus(Z = 0 mm) and away from the focus (Z = +10 mm). Figurs7 (b) and 8 (b) show that the spot of the transmitted beam has minimum size only when the sample was at the focus, Figurs 7(c) and 8 (c) show the spatial ring pattern (self-diffraction).

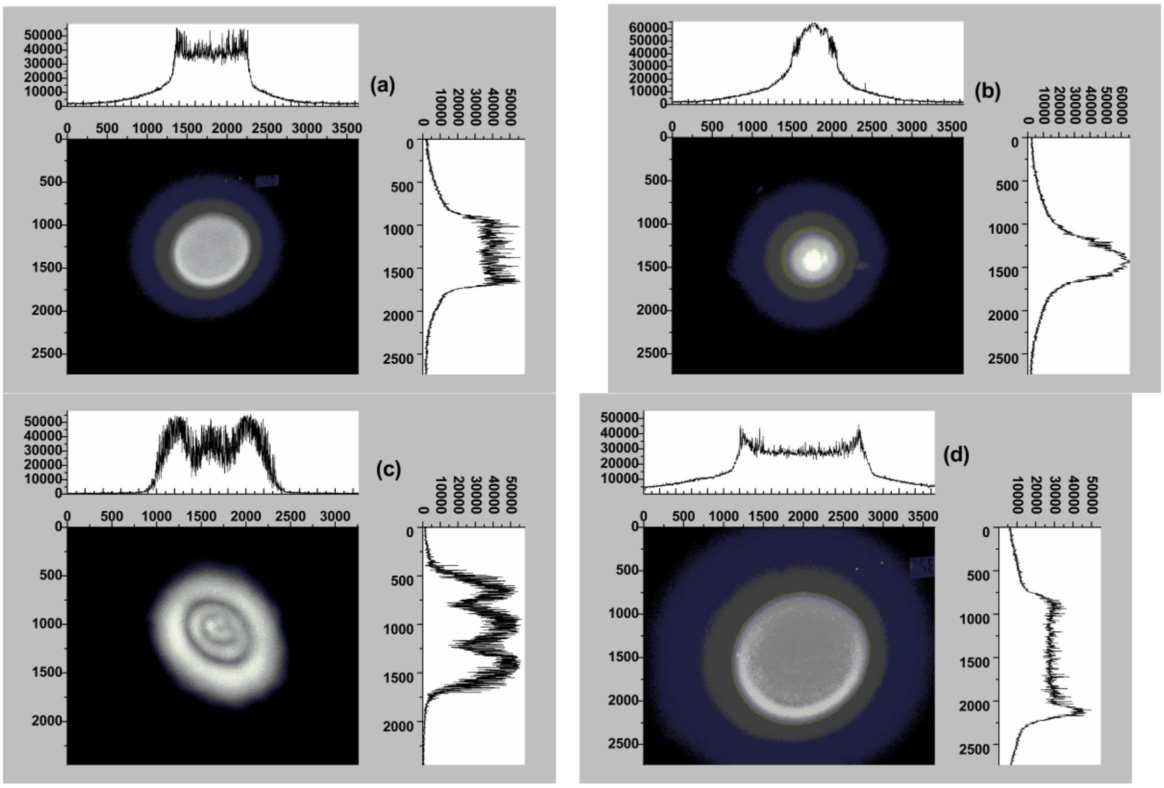

Figure 7. The transmitted beam profiles and the distribution of intensity corresponding to the sample positions :(a) far from the focus (b) at the focus (c) out of focus and (d) away from the focus for pure azo dye solution.

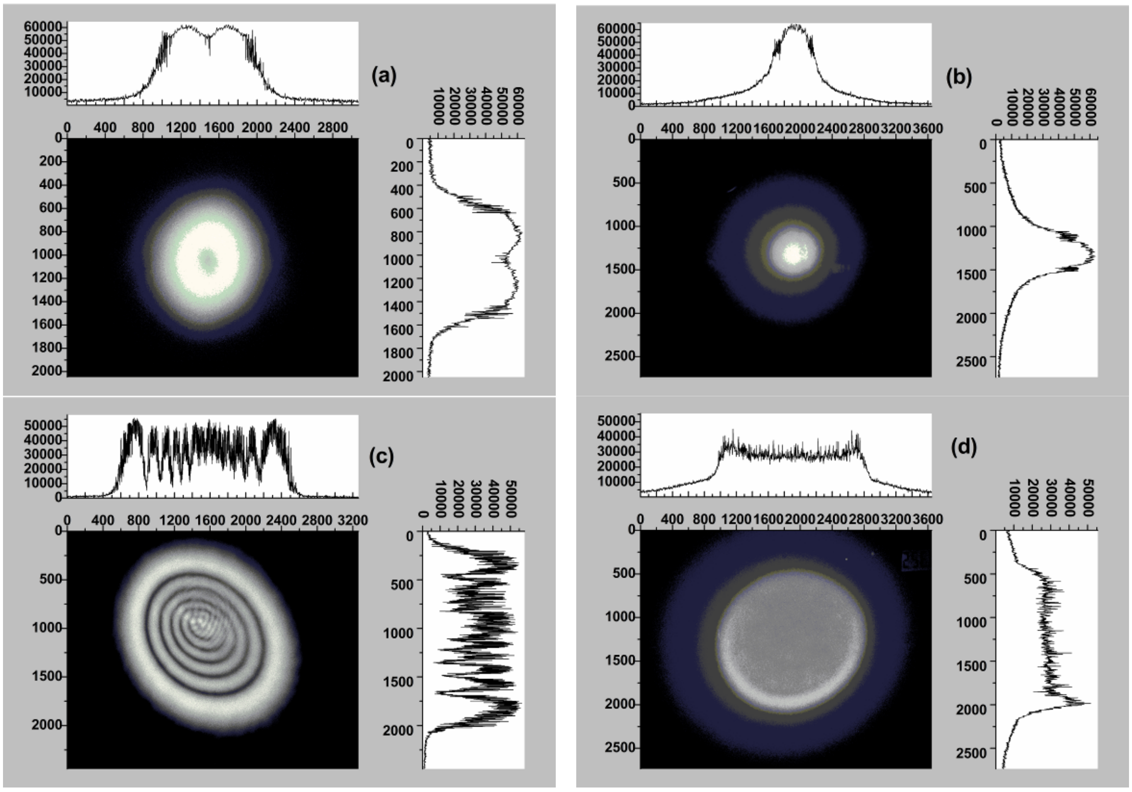

Figure 8. The transmitted beam profiles and the distribution of intensity corresponding to the sample positions :(a) far from the focus (b) at the focus (c) out of focus and (d) away from the focus for azo dye with VO^{+2} solution.

3. Diffraction Ring Techniques

When a laser beam acts on a nonlinear medium, the reasons for which the refractive index change are various and the mechanisms behind the changes in the refractive index are not completely the same (Qimin *et al*.1995). For the photorefractive effect, the change in the refractive index of the nonlinear media has nothing to do with the light intensity, and the light intensity affects merely the speed of the photorefractive process, while for the optical Kerr effect, the change in the refractive index is proportional to the light intensity.

The relationship between $\Delta\varphi$ and number of rings, N, can be written as (Kamikawachi *et al*. 2008):

$$\Delta\varphi_0 = 2\pi N \tag{5}$$

The relationship between the total refractive index , n , and nonlinear part of the refractive index , n_2 , can be written as follows (Villafranca & Saravanamuttu2009):

$$n = n_0 + n_2 I_0 / 2 \quad \text{and} \quad n = n_0 + \Delta n \tag{6}$$

Where n_0 is the background refractive index.

For the thermal nonlinearity and steady state case, the change nonlinear index , Δn , can be expressed as (Callen *et al*. 1967):

$$\Delta n = \frac{dn}{dT}.\frac{I_0 \alpha \omega_\circ^2}{4K} \tag{7}$$

P is the laser input power (P=30 mW) , dn/dT and K are the sample temperature coefficient of refractive index and thermal conductivity (K =0.1567 W/mK) , respectively.

By the combination of equations (3-7) one can calculate, n_2 and dn/dT . The diffraction Ring 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 21.63 μm, the change nonlinear index , Δn , are 0.79×10^{-4} and 2.08 $\times10^{-4}$, respectively and the relative plane distortion , $\Delta\varphi_0$, are measured to be 0.328 and 0.657, respectively. A 1mm wide optical cell containing the solutions of azo dye (pure azo dye and azo dye with VO^{+2}) is translated across the focal region along the axial direction that is the direction of the propagation laser beam. A semitransparent screen of 30 cm × 30 cm, a digital CCD camera and a detector to measure input power. The output of the CCD camera was fed into a computer for further analysis.

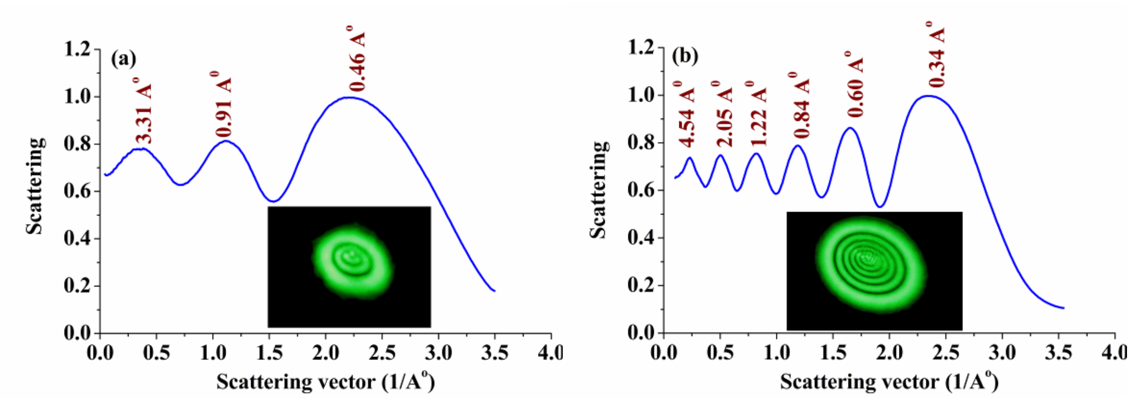

Figure 9. diffraction patterns intensity profile and D- distribution for the sample (a) 3 rings for pure azo dye, (b) 6 rings for azo dye with VO^{+2}.

Figure 9 shows the diffraction patterns intensity profile and D- distribution for the azo dye. The number of observed ring increases in azo dye with vanadyle ion more than pure azo with same input power. In our experiment we obtained a maximum ring number of 6 at an input power of 30 mW. Our patterns are quite concentric and sharp. This is because the scattering of a laser beam in solvent, which is caused by the fluctuation of molecular axes. The outermost ring is the strongest of all the rings and is especially wide. We were not able to obtain definite rings at input power levels over that value, probably due to the boil of (1.8-Dihydroxy-naphthalin-3, 6 disulfonic acid-[2-(4-Azo)]- N - (5-methyl – 3 – isoxazolyl)- benzene sulfonamide) dye in the solvent Dimethyl sulfoxide. As given in Table 2, the nonlinear refractive index , n_2 and thermo-optic coefficient , dn/dT , increases in azo dye with VO^{+2} more than pure azo dye at 0.08 mM concentration.

Table 2. Number of rings, nonlinear refractive index and thermo-optic coefficient

Sample	N	n_2 $\times 10^{-8}$ (cm^2/W)	dn/dT $\times 10^{-6}$ (k^{-1})
Azo dye	3	3.262	1.635
Azo dye+ VO^{+2}	6	8.495	3.126

Conclusion

The linear absorption and nonlinear refraction indices for azo dye (1.8-Dihydroxy-naphthalin-3, 6 disulfonic acid-[2-(4-Azo)]-N-(5-methyl-3-isoxazolyl)-benzene sulfonamide) solution were measured using open-and closed- aperture z-scan techniques, with cw irradiation. The closed aperture Z-scan experiments for sample shows peak–valley characteristic and it is concluded that thermal self defocusing is the most probable mechanism of nonlinearities in this sample and sign of nonlinear refraction is negative. Furthermore, diffraction rings pattern as a result of nonlinear refraction was observed. The diffraction patterns intensity profile and D-distribution for the azo dye are studied experimentally. The number of observed ring increases in dye with vanadyle ion (VO^{+2}) more than pure azo dye with same input power. In our experiment we obtained a maximum ring number of 6 for azo dye with vanadyle ion (VO^{+2}) and 3 for pure azo dye at an input power of 30 mW. Thermal effects appeared to enhance nonlinearities in azo dye under pumping with low power, continuous waveform light from a solid-state diode laser. These effects appeared in the shape of diffraction rings that increased in number with the azo dye with vanadyle ion (VO^{+2}). The increase in number of rings is nonlinear. In the solution samples, due to the thermally induced mechanism, relaxation like behaviour for nonlinearity was observed.

Acknowledgements

The author would like to thank Dr. Alaa yassein Taha Al-Ahmad for his valuable discussions, intellectual support and expertise in LabView programming.

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