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Self diffraction and nonlinear optical properties for 2, 3-Diaminopyridine under cw illumination

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***Abstract:* -** The nonlinear absorption and refraction indices for 2,3-Diaminopyridine solution were measured using open-and closed- aperture z-scan techniques, with continuous wave (cw) irradiation. Furthermore, diffraction rings pattern as a result of nonlinear refraction was observed. The effect of concentration, wavelength and laser intensity on the nonlinear absorption, nonlinear refraction and diffraction rings are studied experimentally. It is found that the nonlinear refraction and absorption indexes in order of 10^{-8} cm^2/W and10^{-3} cm/W, respectively. We suggested an opportunity to form a new nonlinear-optical media for nonlinear optical application.

Keywords:* *- nonlinear refractive index, nonlinear materials, Z-scan, self- diffraction.

I. INTRODUCTION

Nonlinear optics (NLO) is a branch of optics that is associated with the changes in the optical properties of material when it interacts with light. Nonlinear optical materials have been explored greatly for their various applications in all-optical switches, opto-electronic devices, 3-D optical memory devices, optical modulation, tele-communications, human eyes and optical sensors protection, etc., and future applications in biological and medical sciences [1-6]. Continuous wave lasers ranging from mW to kW are widely used in many applications[7]. Wide range of materials including liquid crystals, porphyrins, dyes, semiconductor nanoparticles, thin films, phthalocyanines and crystals are known to be optically nonlinear under cw laser illumination [8-16].

Several techniques developed to measure the nonlinear optical properties such as nonlinear interferometry, degenerate four-wave mixing, nearly degenerate three-wave mixing, ellipse [17,18]. rotation and beam-distortion are sensitive but usually require complex experimental apparatuses The Z-scan technique is a popular and powerful method for the measurements of the optical nonlinearity because of its sensitivity, simplicity and ability to determinate the signs and magnitudes of optical nonlinearity. This method allows the simultaneous measurement of both nonlinear refractive index and nonlinear absorption coefficient. Basically, the method consists of translating a sample through the focus of a Gaussian beam and monitoring the changes in the far field intensity pattern. Because of the light-induced lens-like effect, the sample has the tendency to recollimating or defocusing the incident beam, depending on its z position with respect to the focal plane. By properly monitoring the transmittance change through a small aperture placed at the far-field position (closed aperture), one is able to determine the amplitude of the phase shift. By moving the sample through the focus and without placing an aperture at the detector (open aperture), one can measure the intensity dependent absorption as a change of transmittance through the sample.

In this study, we report the experimental investigation of third-order optical nonlinearity of 2,3-Diaminopyridine by using the single beam Z-scan technique. We also investigated the self-diffraction for the sample under cw laser illumination. The experiment is performed for different concentrations, wavelengths and incident beam intensities. The sample is found to exhibit a negative and large optical nonlinearity.

II. EXPERIMENTAL

The sample and DMSO are purchased from Aldrich Chemical Company and were used without any purification. The chemical structure and molecular formula of 2,3-Diaminopyridine are shown in Fig.1. A UV–visible spectroscopy has been used to characterize the 2,3-Diaminopyridine in solvent DMSO in the spectral range (350–900 nm). The absorbance (A) of the sample measured using Cecil Reflected-Scan CE 3055 reflectance spectrometer. These measured were performed at room temperature. Fig.1 shows the spectral distribution of absorbance of samples with different concentrations. We can see from the Fig. 1 that the absorbance of the sample increases with increasing the concentration this due to increase number of molecular per unit volume, so the absorbance will be increased.

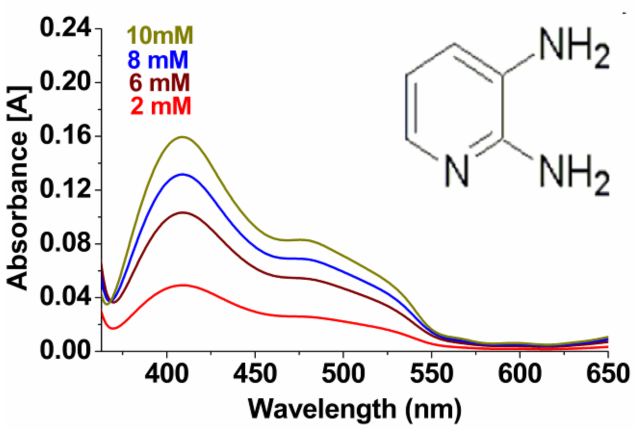

Fig. 1. UV-Visible absorption spectrum of 2,3-Diaminopyridine with different concentrations. Inset shows the chemical structure of 2,3-Diaminopyridine .

The nonlinear absorption and nonlinear refraction of the sample were measured by using the open-and closed-aperture Z-scan technique. The schematic diagram of Z-scan experiment is shown in Fig.2. A cw laser beam from solid state laser SDL at 473 nm wavelength with an average power of 4.6 mW was used as a source of light in our experiment. The output of the laser beam had a Gaussian intensity profile. The Gaussian laser beam was focused by using a lens of focal length 5 cm, into the sample solution contained in a 1 mm quartz cuvette. The resulting beam waist radius at the focus was 22.19 µm and the diffraction length, Z_R was found to be 3.2 mm. The sample was moved along a z-axis by a motorized translational stage. The transmission of the beam through an aperture placed in the far field is measured using photo detector fed to the digital power meter (Field master Gs-coherent). For an open aperture Z-scan, a lens was used to collect the entire laser beam transmitted through the sample with the aperture replaced. The sample thickness of 1 mm was less than the Rayleigh length and hence it could be treated as a 'thin medium'. The measurements were done at room temperature.

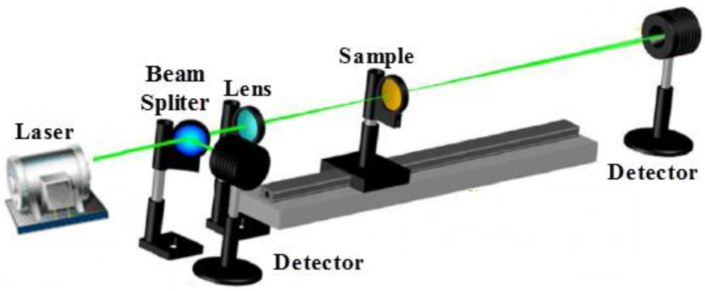

Fig.2. Schematic diagram of experimental arrangement for the Z-scan measurement

III. RESULTS AND DISCUSSION

3-1 THE ABSORPTION COEFFICIENT (A)

The spectrum of the optical absorption was computed from the absorbance data. The absorption coefficient (α) has been obtained directly from the absorbance against wavelength curves using the relation [19]:

$$\alpha = 2.303A/d \qquad \ldots\ldots(1)$$

Where d is the sample thickness and A is the absorbance.

The values of absorption coefficient at wavelength 473 nm for 2,3-Diaminopyridine with different concentrations have been calculated using Eq.1 and they are given in Table 1.

3.2 Nonlinear optical properties

The third order nonlinear optical properties of the sample is studied by conducting the Z-scan experiment. The model described in [20] was used to determine the magnitude of nonlinear absorption coefficient β and the nonlinear refractive index n_2 of the 2,3-Diaminopyridine in solvent DMSO.

The magnitude and sign of nonlinear absorption coefficient, β, of the 2,3-Diaminopyridine was determined through open aperture Z-scan. Fig.3 shows the Z-scan data fo 2,3-Diaminopyridine in solvent DMSO at four different concentration 2,6,8 and 10 mM at incident intensity I_0 = 0.595 kW/cm^2. It can be seen from Fig. 4 that the normalized transmittance decreases when sample moves nearer to the focal point. This is an indication of nonlinear absorption. The nonlinear absorption also increases considerably with the concentration.

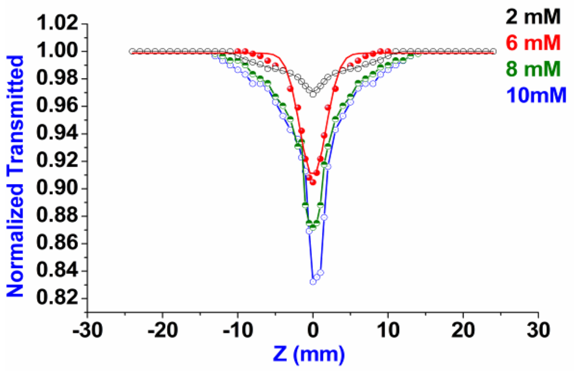

Fig. 3. Open aperture Z-scan data for different concentrations.

The nonlinear absorption coefficient ,β, can be calculated using the equation [21].

$$\beta = \frac{2\sqrt{2}\Delta T}{I_{\circ} L_{eff}} \qquad \text{……………………………………… (1)}$$

Where ΔT is one-valley transmission, I_o is the intensity of the laser beam at focus and L_{eff} =[1-exp(-αL)] /α is the effective thickness of the samples, L is the thickness of the sample.

The closed aperture Z-scan experiments were performed by placing the aperture in front of the detector, which allowed us to determine the sign and magnitude of the nonlinear refractive index,n_2, of sample. The sensitivity to nonlinear refraction is entirely due to aperture, and absence of aperture completely eliminates the effect. Fig. 4 illustrates the closed aperture Z-scan profiles of the samples. The normalized closed aperture Z-scan curve exhibits a pre-focal transmittance maximum (peak) followed by a post-focal transmittance minimum (valley) signature for the samples. This peak-valley signature indicates the self-defocusing property and it is represented by negative nonlinear refractive index n_2. The sign of the nonlinear index of refraction n_2 of a sample is thus immediately clear from the shape of graph.

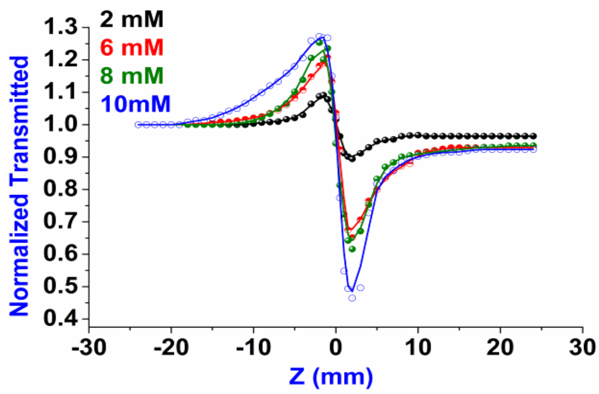

Fig. 4. Closed-aperture Z-scan data for different concentrations.

Self-focusing and self-defocusing were also observed with naked eye. Fig. 6 shows the variation of the spot size at the far-field as a function of the sample position relative to the focal plane for sample. The first photograph (Fig. 6(a)) was taken well before (-z) where the nonlinear effect is not present because of low intensity in this region. Similar spot size and shape was observed long after (+z) (Fig. 6(d)). Fig.6 (b) was taken at the pre-focal transmittance maximum and Fig. 6(c) was taken at the post-focal transmittance minimum. The above implies that self- focusing and self-defocusing can be easily observed with a low-power cw laser. This can be used as a quick check for nonlinear behavior of materials before proceeding with the experiment.

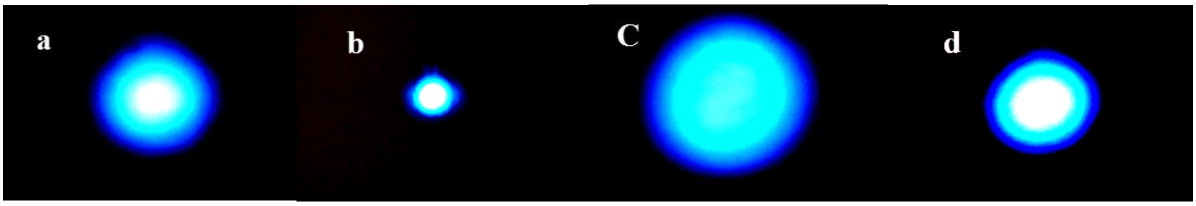

Fig.5. Photographs show the variation of the spot size as a function of the sample position relative to the lens focal point. (a) Before focus, where no nonlinear effects are present. (b) Pre-focus transmittance maximum. (c) Post-focus transmittance minimum (d) After focus, where no nonlinear effects are present.

Since closed aperture data obtained from Z-scan will contain both nonlinear refraction and nonlinear absorption components, it is necessary to separate the nonlinear absorption components from the nonlinear refraction so as to extract pure nonlinear refraction. In order to differentiate nonlinear refraction from nonlinear absorption, the division method is followed [20]. Fig.6 gives the pure nonlinear refraction curve obtained by dividing closed aperture data by the corresponding open aperture data. The physical origin of nonlinear refraction can be electronic, molecular, electrostrictive or thermal in nature. The nonlinear response is related to thermally induced nonlinear effects because the cw laser normally produces heating effect and the defocusing nonlinearity is due to the laser heating process.

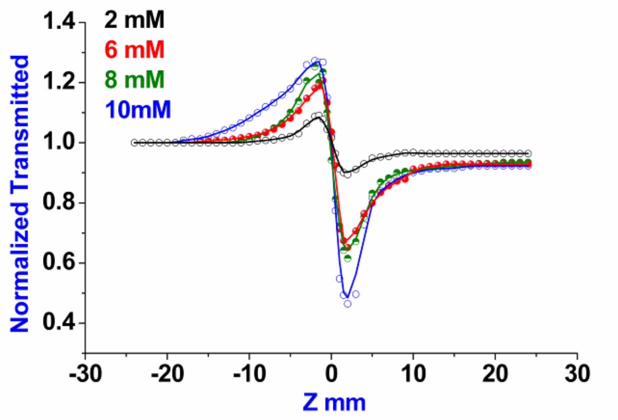

Fig. 6. Pure Z-scan data for different concentrations.

The difference in peak- valley normalized transmittance ΔT_{P-V} can be defined as the difference between the normalized peak and valley transmittances T_P-T_V. The variation of ΔT_{P-V} quantity as a function of $\Delta Ø_o$ is given by [20],

$\Delta T_{P-V} = 0.406\ (1-S)^{0.25}\ \Delta Ø_o$ ……………………………………….. (2)

Where $\Delta Ø_o$ is the on-axis phase shift and $S=1-\exp(-2\ r_0^2/\omega_o^2)$ is the aperture linear transmittance with r_o denoting the aperture radius and ω_o denoting the beam radius at the aperture in the linear regime. The nonlinear refractive index, n_2, is given by

$$n_2 = \frac{\lambda \Delta \varphi_\circ}{2\pi L_{eff} I_\circ}$$ ……………………………………….. (3)

where λ is the laser wavelength.

The estimated values of nonlinear absorption coefficient and nonlinear refractive index of sample solution for different concentrations are given in Table 1.

Table 1: Nonlinear optical parameters for 2,3-Diaminopyridine in the solvent DSMO.

Con. (mM)	α (cm^{-1})	$\Delta\varphi$	β x 10^{-3} (cm/W)	Δn	L_{eff} (mm)	n_2 x10^{-8} (cm^2/W)
2	0.599	0.539	1.53	1.183	0.097	7.037
6	1.258	1.479	4.818	3.349	0.093	19.925
8	1.589	1.179	6.595	4.094	0.092	24.360
10	1.909	2.333	8.761	5.271	0.091	31.361

Fig. 7 shows the Z-scan data for 10 mM concentration of sample solution at different incident beam intensities. The output laser intensities used were 0.219, 0.413 and 0.595 kW/cm^2 respectively. The values of nonlinear absorption coefficient and nonlinear refractive index for sample solution calculated from the Fig. 7a and c and are listed in the Table 2. As expected, the nonlinear features increase when the beam intensity increases.

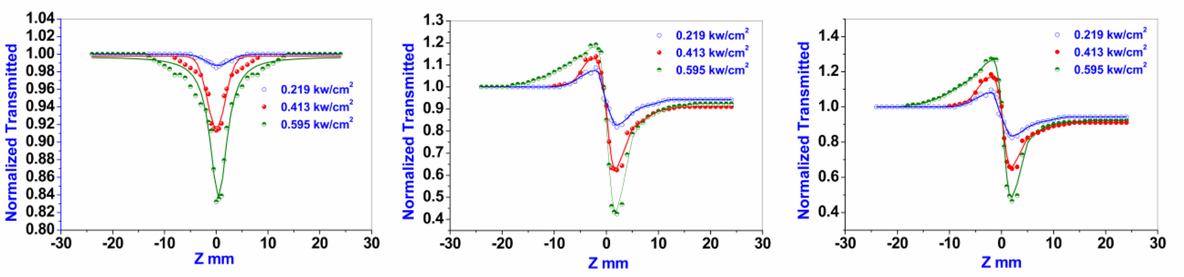

Fig.7. Z-scan curves of the 10 mM solution of 2,3-Diaminopyridine for different incident intensities.

Table 2: Nonlinear optical parameters for different incident intensities.

Intensity (kW/cm^2)	**β x 10^{-3} (cm/W)**	**n_2 x10^{-8} (cm^2/W)**
0.220	0.797	28.510
0.414	4.523	29.898
0.595	8.761	31.361

A study has been made to investigate the influence of the wavelength on the nonlinear absorption coefficient β and nonlinear refractive index for the sample by using the set-up shown in Fig.2. Fig. 8 shows the Z-scan data for open aperture, closed aperture and dividing closed aperture measured at 473and 532 nm wavelengths for a sample solution with 14 mM concentration**.** The Z-scan curves are different for 473 and 532 nm wavelengths as can be seen in Fig.8. The nonlinear absorption coefficient β and nonlinear refractive index of the sample for 473 and 532 nm wavelengths are 33.6x10^{-3} cm/W, 34.816x10^{-8}cm^2/W and 1.073x10^{-3} cm/W, 8.588x10^{-8} cm^2/W, respectively. The deference in amplitude of the Z-scan curves for the two wavelengths can be due to the deference in absorption coefficient at the two wavelengths, where the absorption coefficient α for the sample at 473 nm is 2.3562 cm^{-1},while the absorption coefficient α at 532 nm is 1.2541 cm^{-1}. The output laser intensities used is 0.595 kW/cm^2 for 473 and 532 nm wavelengths.

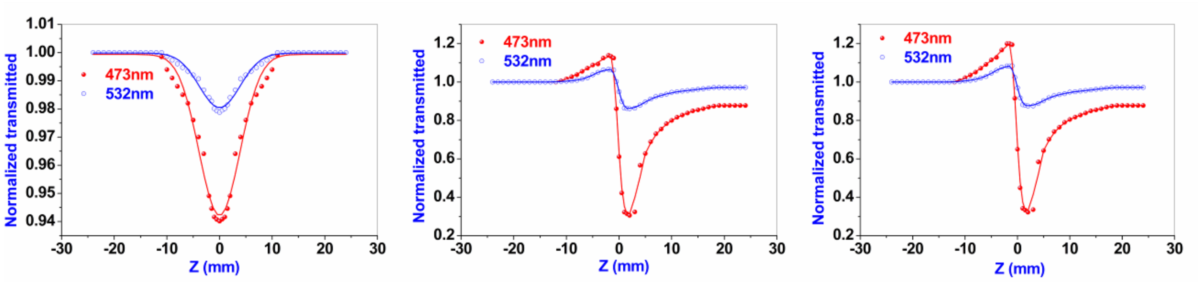

Fig.8. Z-scan curves of the 14 mM solution of 2,3-Diaminopyridine for different wavelengths.

3.3 Self-diffraction pattern

The experimental set-up for the demonstration of self-diffraction of the laser beam by 2,3-Diaminopyridine under cw laser illumination is very similar to the standard Z-scan geometry and the same parameters were used as for the Z-scan experimental set-up. The cuvette containing the nonlinear medium is placed just after the focal point. To observation beam patterns out of the sample we replaced the aperture with an screen 50 cm away from the focal point and were recorded by a digital camera. As the laser power was gradually increased, diffraction rings were observed on the screen when the laser power exceeds a certain value threshold value. The number of rings increases with the increase of laser power. This indicates that they were intensity dependent. A typical pattern of the diffraction rings for incident intensity 1.39, 2.31 and 3.52 kW/cm^2 is shown in Fig. 9. We assumed that thermally induced refractive index change is responsible for the observed diffraction. When the Gaussion beam illuminates the film, the medium absorbs the light and its temperature rises. The rises of temperature results in the change of local refractive index and thus induces the self-diffraction.

Fig.9. Self-diffraction patterns for different incident intensity: (a) 1. 39, (b) 2. 31 and (c) 3.52 kW/cm^2.

The diffraction rings also appear when the sample is illuminated by laser of 473 nm, but there is some difference between the two conditions: (1) the threshold values of the illumination powers for self-diffraction rings are unequal. The threshold value at 473 nm (P_{th} = 1.58 mW) is lower than that at 532 nm (P_{th} = 5.3 mW), which is consistent with the absorption property of sample in this region ; (2) the number of rings varies for different wavelengths under the same intensity. The number of rings at 473 nm is more than that at 532 nm for the same intensity, where the number of rings at 473 nm is 11, while the number of rings at 532 nm is 5 . Fig.10 shows the diffraction rings patterns for the sample at 532 nm and 743 nm wavelength for incident intensity 9.05kW/cm^2.

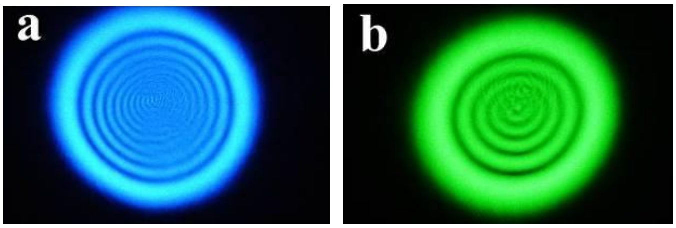

Fig.10. Self-diffraction patterns for different wavelengths: (a) 473 nm (b) 532 nm.

Fig. 11 shows diffraction rings pattern taken for samples with 14,16,18 and 20 mM concentrations. We can see from the Fig. 11 the number of self diffracted rings increases with increasing concentration of the samples. Also the threshold values of the sample with different concentrations for self-diffraction rings are unequal, the threshold values of the sample at 14, 16, 18 and 20 mM concentration are 1.66, 2.35, 3.07 and 3.35 mW respectively. The increasing of the number of diffraction rings with increasing the concentration are due to the increase in aggregation of the sample molecules at the point of focus at higher concentrations. The diffusivity extends to a larger region thereby causing more interference to take place leading to an increased number of rings. For pure DMSO no diffraction patterns were seen. It has been verified that the pure DMSO does not exhibit either self defocusing or self diffraction effects at the power levels used in the experiment.

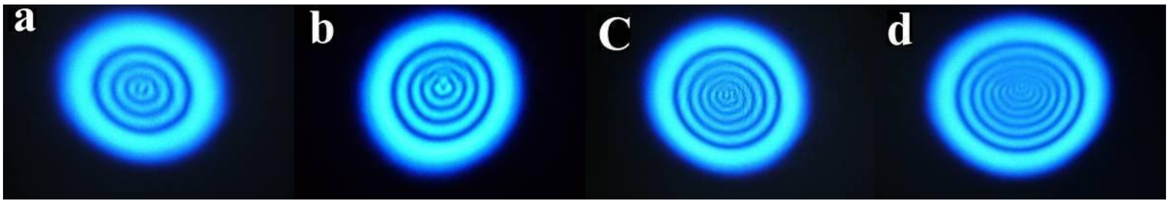

Fig. 11. Self-diffraction patterns at four different concentrations: (a)14 mM, (b)16 mM, (c) 18mM, and (d) 20mM.

IV. CONCLUSION

We have measured the nonlinear refraction index coefficient, n_2, and the nonlinear absorption coefficient, β, for solution of sample using the *Z*-scan technique with 473 nm SDL laser. 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. Moreover, the induced self-diffraction patterns were observed for different concentrations, wavelengths and laser intensities. We think that the sample dissolved in DMSO will be a significant candidate for possible applications in nonlinear optical devices.

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