Thermooptic coefficient and Transmitted beam profile of Rifampin

Hussain Ali Badran, Alyaa A. Jari

Abstract— **The nonlinear optical properties of 3-[[(4-methyl -1-piperazinyl) imino]-methyl] –rifamycin VS (rifampin) in Tetrahydrofuran (THF) are studied by using z-scan and diffraction ring technique with continuous wave (CW) laser at a wavelength of 532 nm. The sample showed negative and large nonlinear refractive index values of the order of 10-7 cm2/W and reverse saturable absorption with high values of the nonlinear absorption coefficient of the order of 10-3 cm/W , and the thermooptic coefficients, dn/dT are found to be of the order of 10-6 K-1. The nonlinear refractive index was found to vary with the concentration. 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 rifampin samples have been studied. These results indicate that the sample is a promising candidate for applications in nonlinear optical field.**

Index Terms— **Nonlinear refractive index, Thermooptic coefficient, Nonlinear absorption, Diffraction ring.**

I. 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 [1]. 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 [2-5]. Among the promising class of materials, organic dyes [6–9] play a vital role because of their good photo-thermal stability, dissolvability etc. The high nonlinear optical refractive index compares favourabl with that of some representative of third-order nonlinear optical materials, namely, CS2 [10], benzo congo red dye solution [11], oxazine (OX720) and oxazine (OX750) dyes in aqueous solution and in polyacrylamide hydrogel (PAA) matrix [10], photopolymerizable organosiloxane [12], and organic polymers [13]. Its potential application is to work as novel optical limiter for its nonlinear optics effect [14-16]. The extensive use of continuous wave lasers for various applications with power levels ranging from W to kW has induced a need to protect the human eyes and sensors [17-22]. 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 .

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We in the present work are presenting the study of nonlinear optical properties i.e. estimation of the nonlinear refractive index, n_2, in rifampin via diffraction ring technique using a visible laser beam. The chosen sample is shown in Fig. 1. We report the results of the refractive nonlinearities studied by using the single beam Z-scan technique on low-cost of rifampin in Tetrahydrofuran (THF) solvent in their resonant region using a continuous wave 532 nm diode- pumped laser. To our knowledge, there is no previous work on the mentioned materials. The aim of our present work is to find the possibility of new applications of the rifampin in the field of the optical modulators. We present experimental evidences of observing diffraction patterns in rifampin at different concentrations with the calculations of the effective nonlinear refractive indices, n_2, and variation of refractive indices.

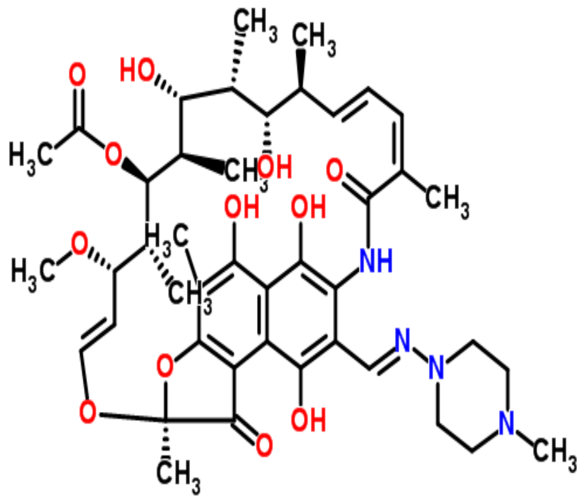

Figure 1. Chemical structure of rifampin

II. EXPERIMENTAL

A. *UV–visible Spectroscopic studies*

In order to study the effect of different concentrations on nonlinear optical properties a UV–visible spectroscopy has been used to characterize the rifampin in the spectral range (225–700nm). The absorbance (A) spectra of solution samples with different concentrations are shown in Figs. 2 measured using Cecil Reflected-Scan CE 3055 reflectance spectrometer. These measured was performed at room temperature. We can see from the Fig.2 that the absorbance of the sample increases with increasing the concentration of the sample this due to increase number of molecular per unit volume, so the absorbance will be increased.

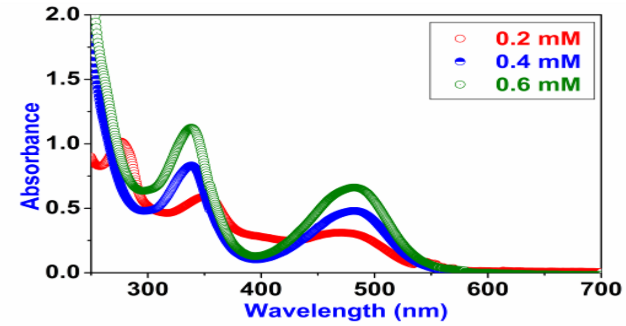

Figure 2. UV-VIS absorption spectrum of rifampin at different concentrations.

B. Z-scan measurement

The Z-scan technique [23,24] 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 Fig.3. A 1 mm quartz cell containing the rifampin solution was translated across the focus of the lens along the direction of the propagation of laser beam.

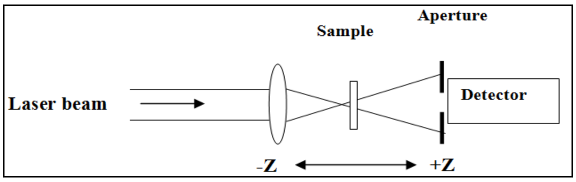

Figure 3. Schematic diagram of experimental arrangement for the Z-scan measurement.

A beam from continuous SDL laser operating at 532 nm and power of 3.34 mW is used to perform the measurement. The beam was focused on the sample using +5 cm focal length lens. Fig. 4 shows the closed aperture Z-scan data of pure sample in THF solvent at different concentrations (0.2 , 0.4 and 0.6 mM). The laser beam waist ω_o at the focus was measured to be 27.04 μm and the Rayleigh length Z_R = 4.31 mm. Thus the sample thickness of 1 mm is less than the Rayleigh length and thus it could be treated as a thin medium. The intensity at the focus is as I_o = 0.29 kW/cm^2.

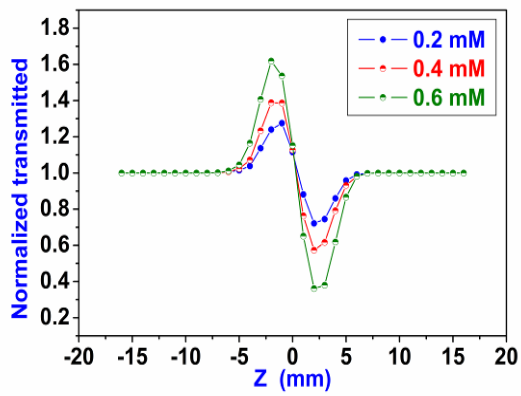

Figure 4. Closed aperture Z-Scan data for rifampin solution.

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. Fig.5 shows the measured Z-scan data for open aperture set-up for the pure rifampin in THF solvent at different concentrations.

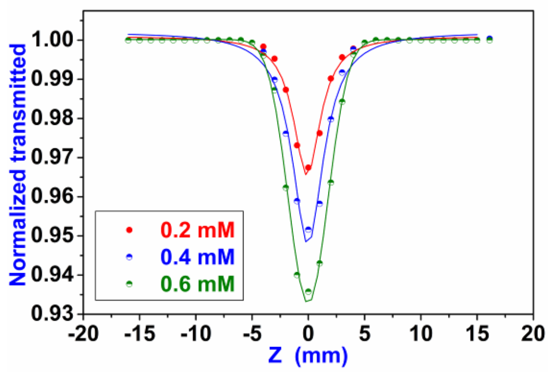

Figure 5. Open aperture Z-Scan data for rifampin solution.

The nonlinear absorption data obtained under the conditions used in this study can be well described by Eq.1 [25-27] which describes a third-order nonlinear absorptive process,

$$\beta = \frac{2\sqrt{2}\Delta T}{I_0 L_{eff}} \qquad (1)$$

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 ($n_2 < 0$) [28]. 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 [28,29]. An easily 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 $\Delta\varphi_0$ is given by[30]:

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

where S is the linear transmission of the aperture and is given by [32]:

$$S = 1-\exp(-2r_a^2/(\omega_a^2)) \qquad (3)$$

where r_a = 2.5 mm is the radius of the aperture and ω_a =6.28 mm is a 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 [32]:

$$\Delta\varphi_0 = (2\pi L_{eff} n_2 I_0)/\lambda \qquad (4)$$

where $k = 2\pi/\lambda$ is the wave vector in vacuum and λ is the laser beam wavelength, I_0 is the intensity of the laser beam at focus z= 0, 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 Figs. 4 and 5 scans is shown in Fig.6. The values of the linear absorption and nonlinear refractive index of rifampin in THF solvent at different concentrations are given in Table1.

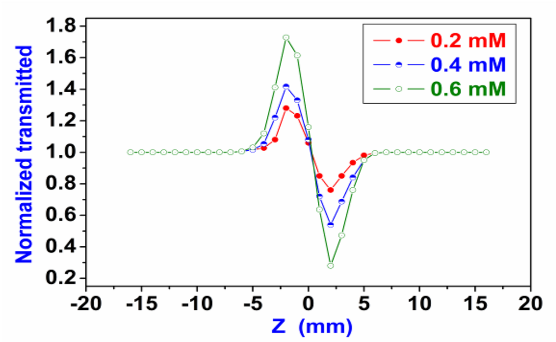

Figure 6. Pure nonlinear refraction curve for rifampin solution.

In Z-scan measurement, the transmittance of the sample measured without an aperture gives information on purely nonlinear absorption coefficient where as the apertured scan contains the information of both the nonlinear absorption coefficient and nonlinear refractive index nonlinearities. The ratio of the normalized closed aperture and open aperture scans generates a Z-scan due to the purely nonlinear refractive index [33-36] and results are shown in Fig.6.

Table1. The linear absorption coefficient and nonlinear optical parameters.

Con. (mM)	α cm^{-1}	$\Delta\varphi_0$	n_2 $\times10^{-7}$ (cm^2/W)	$dn/dT\times10^{-5}$ K^{-1}	$\beta\times10^{-3}$ (cm^2/W)
0.2	1.62	1.41	4.45	18.83	3.42
0.4	2.54	2.38	7.87	21.27	5.33
0.6	3.58	3.93	13.63	26.21	7.43

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 2π, a set of concentric rings will appear on the far-field observation screen as the Gaussian beam is transmitted through the nonlinear medium [37]. The spot of the transmitted beam was photographed at far away distance from the sample, when the sample was at different positions. Fig. 7(a)–(d) shows the distribution of intensity for rifampin in THF solvent at 0.4 mM, 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). Fig. 7 (b) shows that the spot of the transmitted beam has minimum size only when the sample was at the focus, Fig. 7 (c) shows the spatial rings pattern (self-diffraction).

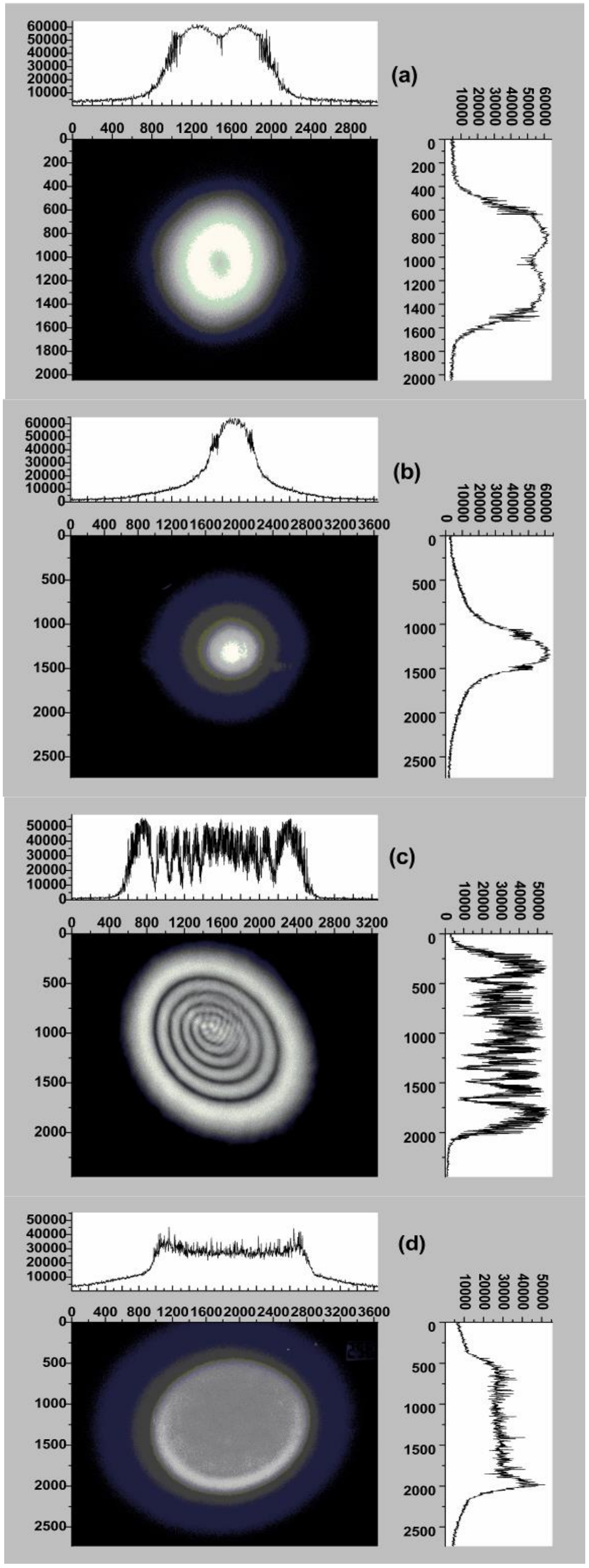

Figure 7. The transmitted beam profiles and the distribution of intensity corresponding to the sample positions: (a) far from the focus (b) at the focus (c) out of focus and (d) away from the focus .

III. 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 [38]. 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 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, Δn, 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, $\Delta\varphi$, suffered by the beam while traversing the sample of thickness (L) can be written as [39]:

$$\Delta\varphi = kL\Delta n \quad (5)$$

where k=2ππ/λλis the wave vector in vacuum and λλis the laser beam wavelength. The relationship between $\Delta\varphi$ and number of rings, N, can be written as [40]:

$$\Delta\varphi = 2\pi N \quad (6)$$

The relationship between the total refractive index , n , and nonlinear part of the refractive index , n_2 , can be written as follows [41,42]:

$$n = n_0 + n_2 I_0 / 2 \text{ and } n = n_0 + \Delta n \quad (7)$$

Where n_0 is the background refractive index. A 1mm wide optical cell containing the solutions of rifampin in THF solvent 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. By the combination of equations (5-7) one can calculate nonlinear refractive index, n_2 . 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 27.04 μm and the input laser power is 25 mW. The change nonlinear index, Δn and the nonlinear refractive index are given in table 2. As given in table 2, the number of rings N for 25 mW power, observed are 5, 7 and 9 respectively. The diffraction rings pattern for the rifampin in THF solvent are shown in Fig. 6.

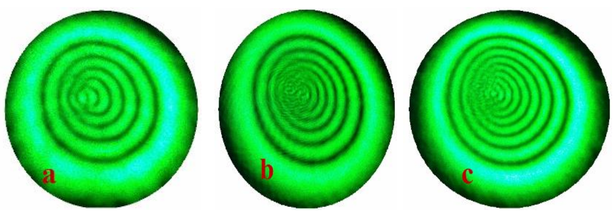

Figure 6. diffraction ring patterns for the rifampin at 25 mW (a) 5 rings for 0.2 mM (b) 7 rings for 0.4 mM and (c) 9 rings for 0.6 mM.

Table 2. Number of rings, nonlinear refractive index and the change nonlinear index.

Con. (mM)	N	$n_2 \times 10^{-7}$ cm^2/W)	Δn $\times 10^{-4}$
0.2	5	0.230	0.502
0.4	7	0.337	0.735
0.6	9	0.458	0.998

IV. CONCLUSION

The linear absorption and nonlinear refraction indices for 3-[[(4-methyl-1-piperazinyl) imino]-methyl] –rifamycin VS in THF solvent 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. We have measured the nonlinear refraction index n_2 and the nonlinear absorption coefficient,β, for the solutions of rifampin for various concentrations using the Z-scan technique with 532 nm wavelength. The Z-scan measurements indicated that the sample 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 sample. All the solutions samples showed a large nonlinear refractive index of the order of 10^{-7} cm^2/W and 10^{-3}cm/W, respectively. Furthermore, diffraction rings pattern as a result of nonlinear refraction was observed. The diffraction patterns were found to vary with the concentrations in our experiment. Experimental results of ring patterns suggest the possibility of using rifampin for various concentrations solvent in THF in all optical systems. These profiles demonstrate that a bright diffraction ring gradually becomes thicker from inner to outer side, and the light energy is mainly concentrated inside the outermost ring. Such behavior corresponds to that observed earlier for divergent Gaussian beams passing through self-defocusing media. Notice that, in general, self-defocusing media have a negative optical nonlinear birefringence Δn [43,44]. The number of rings depends on the concentration, that is, increasing the concentrations values increases the number of rings for the same input power. This means that, in the investigated samples, thermal effects have a large contribution to the negative nonlinear refractive index. The heat released in the rifampin solution by the focused Gaussian laser beam causes a migration of the solutes in the different concentrations from the hotter region to the colder one. All these experimental results show that the solutions of rifampin are a promising material for applications in nonlinear optical devices.

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