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MEASUREMENTS OF THIRD-ORDER SUSCEPTIBILITY OF 3-[[(4-METHYL-1-PIPERAZINYL) IMINO]-METHYL]- RIFAMYCIN VS USING Z-SCAN TECHNIQUE

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ABSTRACT

The nonlinear optical properties of 3-[[(4-methyl-1-piperazinyl) imino]-methyl]-rifamycin VS in Dimethyl sulfoxide (DMSO) solvent with different concentrations were studied using single beam Z-scan technique with a continuous-wave radiation at 532 nm of an output power of 3.34 mW. The Z-scan measurements indicated that the samples exhibited large nonlinear optical properties. The optical response was characterized by measuring the concentration dependent refractive index n_2 *of the medium using the Z-scan technique. The sample showed negative and large nonlinear refractive index values of the order of* 10^{-7} cm^2/W *and reverse saturable absorption with high values of the nonlinear absorption coefficient of the order of* 10^{-3} *cm/W. The origin of optical nonlinearity in the samples may be attributed due to laser-heating induced nonlinear effect. These results indicate that the sample is a promising candidate for applications in the nonlinear optic field.*

KEY WORDS-Nonlinear optics, Nonlinear refractive index, Z-scan, self-defocusing.

INTRODUCTION

Materials with third-order optical nonlinearities have been investigated extensively for their applications in optical switching, optical phase conjugation, optical limiting devices, etc. It has been observed from the reported literatures and some theories that organic compounds have large third-order NLO susceptibility $\chi^{(3)}$ value and fast response, [1-14] and are promising candidates for these nonlinear optical applications [15,16]. The nonlinear sample acts as a focusing lens when its nonlinear refractive index n_2 is positive, but it diverges the incident beam [17] when its nonlinear refractive index n_2 is negative. The satisfying strong nonlinear refraction enables the facility of optical switching, while the presence of strong nonlinear absorption produces good optical limiting [18].

Based on the above consideration, meanwhile, in order to find a new organic compounds and as a continuation of our search for better NLO materials [19-22] we choose 3-[[(4-methyl-1-piperazinyl) imino]-methyl] –rifamycin VS as a sample in this study. In this method a single focused beam is used to illuminate the sample. The transmittance through an aperture placed some distance from the focal plane of the beam is monitored as the sample is scanned along the optic axis. This is called closed-aperture experiment. From this measurement both the sign and magnitude of n_2 can be determined. For materials with a negative n_2 (self-defocusing), the profile of the Z-scan transmittance curve consists of a peak followed by a valley as the sample is translated from –Z to +Z. For positive n_2 materials (self-focusing) the profile is reversed with a valley–peak sequence. In cases in which the material has a nonlinear absorption, the aperture of the Z-scan setup is removed. This is called open-aperture experiment. While the effective nonlinear absorption coefficient can be determined from the open-aperture experiment, the effective nonlinear refraction coefficient can be determined from closed/open-aperture curves for nonlinear absorptive materials [18].

The experimental work was aimed at investigating the nonlinear refractive index and nonlinear absorption coefficient of 3-[[(4-methyl-1-piperazinyl) imino]-methyl]–rifamycin VS under a continuous wave (CW) light from solid state laser at 532 nm wavelength by using Z-scan technique. The negative refractive index and nonlinear absorption coefficient are measured for different concentrations. Also the third-order susceptibility response arising due to self-defocusing. The results are discussed and the implied mechanism is given.

MATERIALS AND METHODS

3- [[(4-methyl-1-piperazinyl) imino]-methyl]–rifamycin VS, from organic dye family was chosen for the study is a semisynthetic antibiotic derivative of rifamycin SV. Rifampin is a red-brown crystalline powder very slightly soluble in water at neutral pH. Its molecular weight is 822.94 g/mol and its chemical formula is $C_{43}H_{58}N_4O_{12}$. The molecular structure of rifamycin SV and elemental composition of $C_{43}H_{58}N_4O_{12}$ are shown in Fig. 1 and table 1.

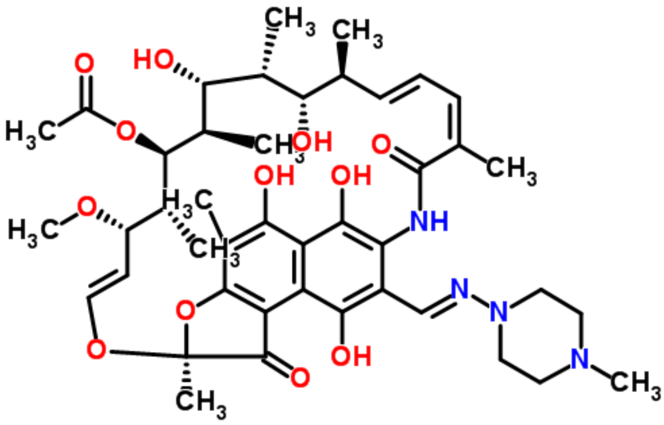

Figure 1: Chemical structure of rifamycin SV.

Table 1: The elemental composition of $C_{43}H_{58}N_4O_{12}$.

Symbol	Element	Atomic weight	Atoms	Mass percent
C	Carbon	12.0107	43	62.7579 %
H	Hydrogen	1.00794	58	7.1039 %
N	Nitrogen	14.0067	4	6.8081 %
O	Oxygen	15.9994	12	23.3301 %

The spectral property (absorption) of the 3-[[(4-methyl-1-piperazinyl) imino]-methyl]-rifamycin VS was dissolved in Dimethyl sulfoxide (DMSO) solvent medium of concentrations 0.1, 0.3, 0.5 and 0.9 mM is studied by recording the absorption spectra of the sample using an UV-VIS spectrophotometer (Cecil Reflected-Scan CE3055 reflectance spectrometer). The absorption spectrum of the sample is shown in Fig. 2.

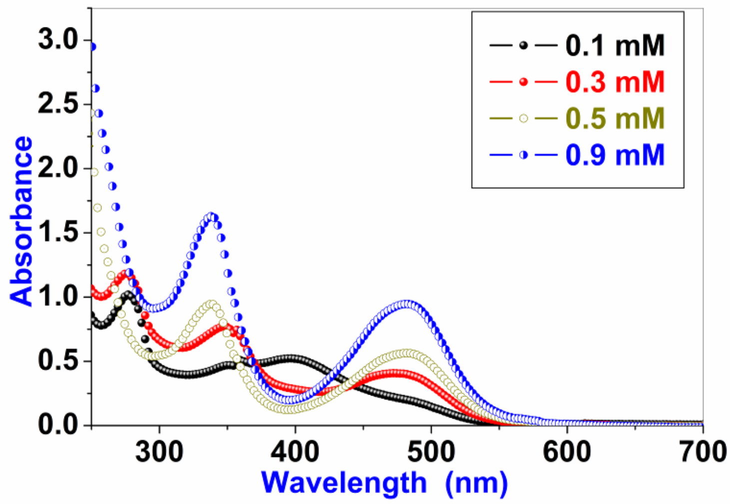

Figure 2: UV-VIS absorption spectrum of rifamycin SV.

NONLINEAR OPTICAL PROPERTIES

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 [23,24]

$$\alpha = 2.303A/d \quad (1)$$

where d is the sample thickness and A is the absorbance. The values of absorption coefficient (α) at wavelength 532 nm for 0.1, 0.3, 0.5 and 0.9 mM concentrations of rifamycin SV in DMSO solvent are 0.13, 1.05, 2.98 and 3.63 cm^{-1}, respectively.

The Z-scan technique is a single-beam method for measuring the optical nonlinearities through detecting the far-field sample transmittance of a focused Gaussian beam as a function of sample position (Z) [25,26]. The schematic of the experimental set up used is shown in Fig. 3. The measurements of third-order nonlinear optical properties of the samples were carried out by using the standard Z-scan method. The excitation source was a continuous wave light from solid state laser (SDL) at 532 nm wavelength, which was focused by 5 cm focal length lens. 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. A 1 mm thick optical cell containing the sample was moved step by step along the propagation direction of the Gaussian beam. The transmission of the beam through an aperture placed in the far field was measured using photo detector fed to the digital power meter. For an open aperture Z-scan, a lens replaced the aperture to collect the entire laser beam transmitted through the sample. Nonlinear refraction and nonlinear absorption were performed by both open and closed-aperture Z-scans of a series of the samples at room temperature [27].

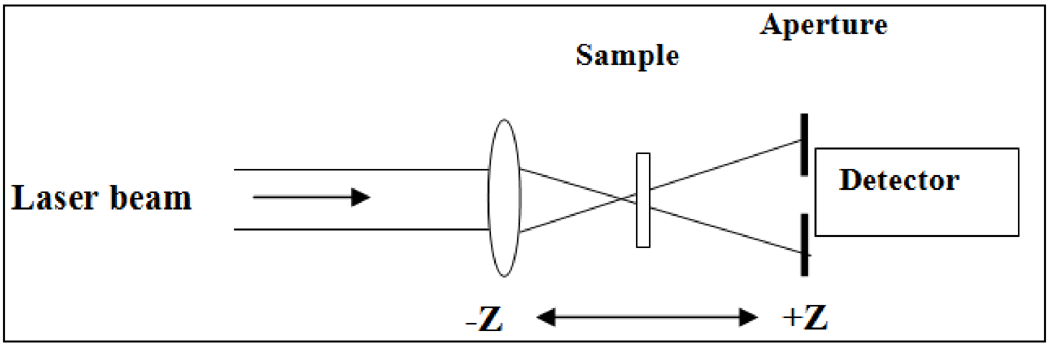

Figure 3: Schematic diagram of experimental arrangement for the Z-scan measurement.

RESULTS AND DISCUSSION

The third-order nonlinear absorption and refraction are investigated by *Z*-scan techniques [28-30] which are simple and sensitive experimental technique for the study of nonlinear optical properties and allow determining the sign of the nonlinear refractive and absorption indices.

Fig. 4 shows the closed aperture Z-scan data for 0.1, 0.3, 0.5 and 0.9 mM concentrations of 3 - [[(4-methyl-1 -piperazinyl) imino]- methyl]- rifamycin VS in DMSO solvent at incident intensity I_0= 0.29 kW/cm^2. The scan of different concentrations have peakvalley configuration, corresponding to a negative nonlinear refraction index i.e. self-defocusing occur. The defocusing effect is attributed to a thermal nonlinearity resulting from the absorption of a tightly focused beam traversing through the absorbing dye medium produces spatial distribution of the temperature in the sample and, consequently, a spatial variation of the refractive index that acts as thermal lens resulting in phase distortion of the propagating beam.

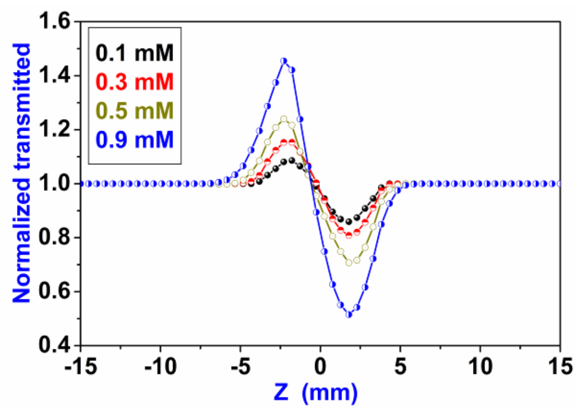

Figure 4: Closed-aperture Z-scan data.

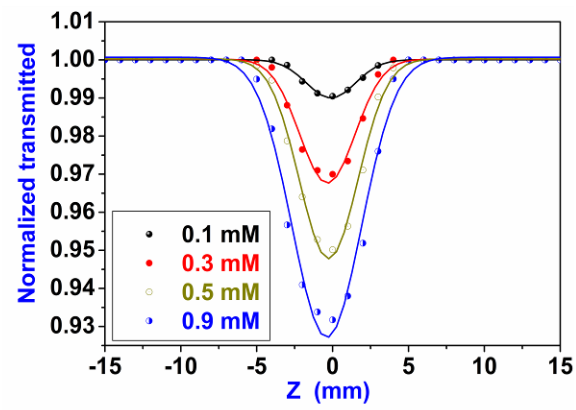

Figure 5: Open-aperture Z-scan data.

The nonlinear absorption component of the rifamycin SV is evaluated under an open aperture configuration. The nonlinear absorption data obtained under the conditions used in this study can be well described by Eq.2 [31-33] which describes a third-order nonlinear absorptive process

$$\beta = \frac{2\sqrt{2}\Delta T}{I_{\circ} L_{eff}}$$

(2)

where ΔT is one-valley transmission for the open aperture, L_{eff} = [1-exp(-αL)]/α is the effective thickness of the sample. Fig. 5 shows the normalized transmittance plotted as a function of the sample position (Z) measured by using an open aperture Z-scan technique. The large dip observed around the focal position is due to the nonlinear absorption. The nonlinear absorption coefficient β (cm/W) for rifamycin SV with different concentrations are calculated from the open aperture normalized transmittance in Fig. 5 and Eq. 2. The values of β are given in Table2.

Fig. 6 shows a typical closed aperture Z-scan experimental curve of the rifamycin SV. The concentrations sample have similar peak-valley pattern of the normalized transmittance curve obtained under the closed aperture configuration indicates that the sample have a negative sign for the nonlinear refraction and exhibits strong self-defocusing behavior. The difference in amplitude of the Z-scan curves for the different concentrations can be due to the difference in absorption coefficient at the 532nm wavelength. In Z-scan measurement, the transmittance of the sample measured without an aperture gives information on purely nonlinear absorption coefficient whereas 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 [29, 34] and results are shown in Fig.6.

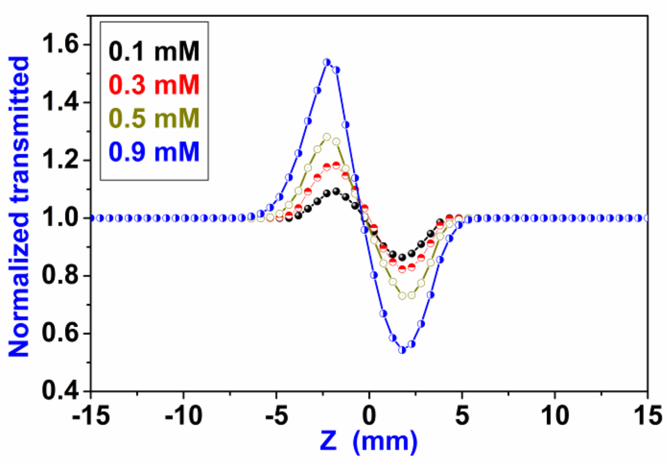

Figure 6 : Pure Z-scan data.

Let $|\Delta\phi_o|$ be the on-axis phase shift at the focus which is related to the difference in the peak and valley

transmission ΔT_{p-v} as [35]:

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

(3)

where S=1- $exp\ (-2r_a^2/\omega_a^2)$ is the aperture linear transmittance with r_a denoting the aperture radius and ω_a denoting the beam radius at the aperture in the linear regime [36]. The nonlinear refractive index is then given by [37].

$$n_2 = \frac{\Delta\phi_\circ \lambda}{2\pi L_{eff} I_\circ}$$

(4)

where λ is the laser wavelength. I_0 is the intensity of the laser at focus z =0, Also, Δn can be related to the total refractive index of the medium n and the background refractive index no, as follows [38].

$n = n_o + \Delta n$ and $\Delta n = n_2 I$.

(5)

The nonlinear refractive index n_2 (cm^2/W) for the different concentrations sample are calculated from the closed aperture normalized transmittance in Fig. 6 and Eq. 4, and the values obtained are given in Table 2.

Using Eqs. 6 and 7 with esu units [23] the nonlinear refraction index n_2 (in cm^2/W) and the nonlinear absorption coefficient β (in cm/W) are proportional to the real $\chi^{(3)}$ (esu) and imaginary $\chi^{(3)}$ (esu) parts of $\chi^{(3)}$ (third-order susceptibility), respectively .

$$\mathrm{Re}\,\chi^{(3)}(esu) = 10^{-4}\frac{\varepsilon_0 c^2 n_0^2}{\pi} n_2 (\frac{cm^2}{W})$$

(6)

and

$$\mathrm{Im}\,\chi^{(3)}(esu) = 10^{-2}\frac{\varepsilon_0 c^2 n_0^2 \lambda}{4\pi^4} \beta(\frac{cm}{W})$$

(7)

Here n_o, ε_o and c and denote the linear refraction index of each concentration, the electric permittivity of free space (8.854 x 10^{-12} F/m) and the velocity of light (3 x 10^8 m/s), respectively. The absolute value of $\chi^{(3)}$ is given by

$$\left|\chi^{(3)}\right| = \left[(\mathrm{Re}(\chi^{(3)}))^2 + (\mathrm{Im}(\chi^{(3)}))^2\right]^{1/2}$$

(8)

The absolute values of $\chi^{(3)}$ for 3-[[(4-methyl-1-piperazinyl) imino]-methyl]–rifamycin VS with different concentrations are calculated using the values of n_2 and β and Eqs. 6 ,7 and 8, and they are given in Table 2.

Table 2: Nonlinear optical parameters and third-order susceptibility for3- [[(4-methyl-1 -piperazinyl) imino] -methyl] –rifamycin VS.

Sample Concentration	α (cm^{-1})	β x 10^{-3} (cm/W)	n_2 x 10^{-7} (cm^2/W)	Δn x 10^{-3}	$\|\chi^{(3)}\|$ x 10^{-6} (esu)
0.1	0.13	0.93	1.92	0.05	8.33
0.3	1.05	3.07	3.20	0.09	14.18
0.5	2.98	5.60	5.43	0.15	25.54
0.9	3.63	7.91	10.08	0.29	48.85

CONCLUSION

In summary, The Z-scan measurements indicated that the 3-[[(4-methyl-1-piperazinyl) imino]- methyl]- rifamycin VS 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. We have measured the nonlinear refraction index coefficient n_2 and the nonlinear absorption coefficient β for solutions of sample for various concentrations using the Z-scan technique with 532 nm cw laser. Using the single beam z-scan technique, the nonlinear refractive index n_2, and the nonlinear absorption coefficient β of 3 - [[(4-methyl-1 -piperazinyl) imino] -methyl] –rifamycin VS were measured. The concentrations sample showed a large nonlinear refractive index and absorption coefficient of the order of 10^{-7} cm^2/W and 10^{-3} cm/W, respectively. The origin of optical nonlinearity observed in the CW regime is attributed to the thermal variation of local refractive index in the medium. The obtained results indicate that the concentrations sample exhibits positive nonlinear saturable absorption (SA) and negative refraction nonlinearity, manifestation of self-defocusing effect. The results indicate that the sample exhibit self-defocusing nonlinearities.

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