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Nonlinear Properties of Rhodamine C Dissolved in 1-Butanol

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Abstract: The research included the study of the spectral properties of Rhodamine C dissolved in 1-butanol in different concentrations ($7*10^{-6}$, $1*10^{-5}$, $3*10^{-5}$, $5*10^{-5}$, $7*10^{-5}$ mol/L) at ambient temperature and studies its nonlinear properties with the calculation of some parameter like "Nonlinear refractive index" and the "Nonlinear absorption coefficient" in addition to select one of these concentrations ($7*10^{-6}$ mol/L) to study the effect of changing the pinhole diameter on some of these parameter.

Key words: Z-scan technique, nonlinear refractive index (n_2), nonlinear absorption coefficient (β), Rhodamine dye, Rayleigh range, absorption spectrum, transmittance spectrum

INTRODUCTION

Recent essential and applied studies focus on materials with nonlinear optical properties (Christodoulides *et al*., 2010). A tremendous variety of materials comprising organic and inorganic semiconductors, dye molecules, etc. (Chan *et al*., 2008; Ivanov *et al*., 2013; Kiran *et al*., 2002; Wang *et al*., 2014; Yelleswarapu *et al*., 2010; Yamashita, 2012; Khoo, 2014) show nonlinear optical properties which can be used in a variety of applications (Haripadmam *et al*., 2014; Four *et al*., 2011; Yelleswarapu *et al*., 2006; Zhou and Wong, 2011; Christenson *et al*., 2014; Andrade *et al*., 2010; Boni *et al*., 2010).

Optical phenomena which are obtained as a result of the medium modification by light existence can be studied by nonlinear optics. Laser light usually can be modified material properties. Organic materials are one of the significant nonlinear optical materials due to their properties like, show large nonlinear properties, rapid nonlinearities response, simple to operates and integrated into optical devices (Alhamdani *et al*., 2006).

Z-scan technique is one of techniques for the measuring "Nonlinear Refraction" (NLR) and "Nonlinear Absorption coefficient" (NLA) in materials which originally suggest by Sheikh-Bahae *et al*. (1990) where its depends on the basics of spatial beam disfigurement and show simplicity in addition to extremely sensitivity for measuring each of "Nonlinear refractive index" and "Nonlinear absorption coefficient" (Sheik-Bahae *et al*., 1989; Van Stryland; Sheik-Bahae, 1998). Many researchers have been studied the nonlinear properties of organic dyes and both (NLR) and (NLA) were reported (Nader, 2017; Al-Hamdani, 2017; Choubey *et al*., 2014; Sreeja *et al*., 2012; Sukumaran and Ramalingam, 2006; Diallo *et al*., 2014). The main objective of the present research is to identify the nonlinear behavior of the Rhodamine C dissolved in 1-butanol in visible region.

MATERIALS AND METHODS

Experimental part: Rhodamine C (Diethyl-m-amino phenolphthalein) which is a member of the xanthene family from (Himedia). India company were dissolved in 1-butanol from "E. Merck AG, Darmstadt Company" (Germany) to prepare different concentrations ($7*10^{-6}$, $1*10^{-5}$, $3*10^{-5}$, $5*10^{-5}$, $7*10^{-5}$ mol/L) at ambient temperature.

Z-scan tests were performed utilizing a (532 nm) "Nd-YAG (SHG) CW" laser beam from (Changchun Company) focused by a lens with focal length equal to (10 cm) and the photo detector type is (S121C) from Changchun Company.

Mathematical equations: The following equations can be used to calculate the values of (ω_0), (Z_R):

$$Z_R = \frac{\pi(\omega_0)^2}{\lambda} \quad (1)$$

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Where:

Z_R = Rayleigh range (mm)
λ = Wavelength of laser (532 nm)
ω_0 = Beam waist and its measured value was (0.0675 mm)

$$\omega(z) = \omega_0 \sqrt{1+[\frac{Z}{Z_R}]^2} \quad (2)$$

Where:

$\omega(z)$ = Half beam diameter (mm)
ω_0 = Beam waist, Z is axial distance
Z_R = Rayleigh Range

The "Nonlinear refractive" (n_2) value was calculated from the equation (Alhamdani *et al*., 2006):

$$n_2 = \frac{\Delta\Phi_O}{I_O L_{eff} K} \quad (3)$$

Where:

$\Delta\Phi_O$ = Nonlinear phase shift
I_0 = Intensity of the laser beam at focus Z = 0
L_{eff} = Effective thickness of sample
k = The wave number

$$\Delta T_{P\text{-}V} = 0.406|\Delta\Phi_O| \quad (4)$$

where, $\Delta T_{p\text{-}v}$ the difference between the normalized peak and valley transmittances:

$$L_{eff} = \frac{(1-e^{-\alpha_o L})}{\alpha_O} \quad (5)$$

Where:

α_o = Linear absorption coefficient
L = Sample thickness

The nonlinear absorption coefficient was calculated from the equation (Alsous *et al*., 2014):

$$T(Z) = \sum_{m=0}^{\infty} \frac{(-q_o)^m}{(m+1)^{\frac{3}{2}}} \quad (6)$$

$$q_o(Z) = \frac{I_O L_{eff}\beta}{1+\frac{Z^2}{Z_O^2}} \quad (7)$$

where, Z the position of sample.

RESULTS AND DISCUSSION

The dye concentration in the solution is proportional to the absorption intensity and it is inversely proportional with the permeability this corresponds to the beer-lambert law. The Full Width at Half Maximum (FWHM) for absorption and transmittance spectrum increases with increasing concentration which effects on the energy state. This is due to the increase in the number of dye particles in the volume unit as shown in Fig. 1 and 2 and Table 1.

Open Z-scan aperture experiments have been used in order to calculate the nonlinear absorption coefficient (β) of different concentrations ($7*10^{-6}$, $1*10^{-5}$, $3*10^{-5}$, $5*10^{-5}$, $7*10^{-5}$ mol/L) from rhodamine C dye dissolved in 1-butanol) Table 2. Figure 3 showed laser beam transmittance after passing through the sample, the nonlinear absorption coefficient of Rhodamine C solutions exhibits the behavior of two-photon absorption and this is due to a change in the intensity of the laser which is moving through the beam waist on the sample.

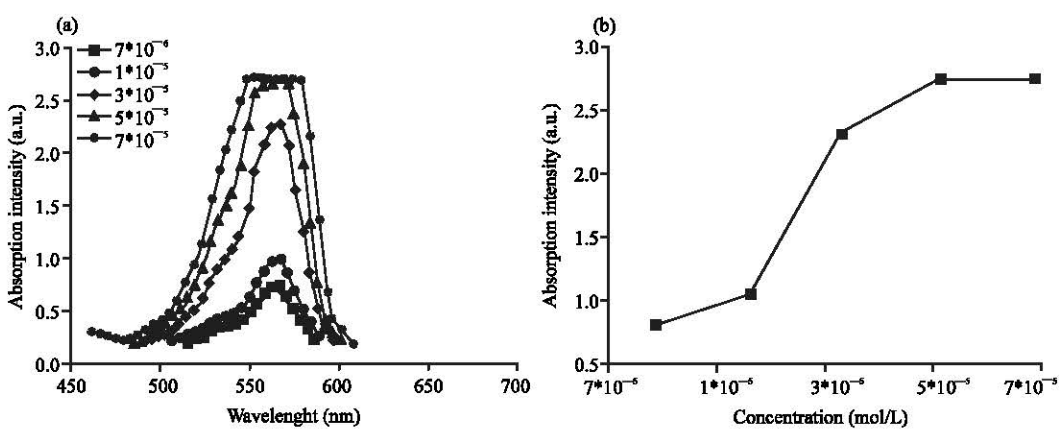

Fig. 1: a) Absorption spectrum of different solutions concentration of Rhodamine C and b) The relation between maximum absorption intensity and different solutions concentration; Maximum value of intensity

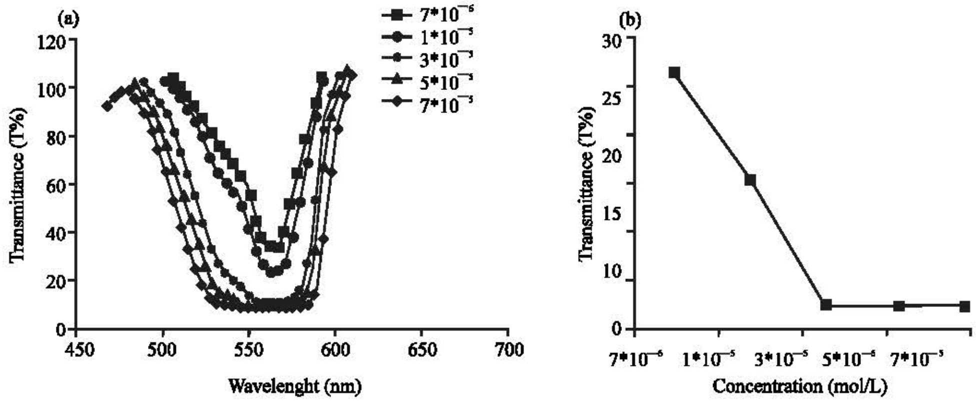

Fig. 2: a) Transmittance spectrum of different solutions concentration of Rhodamine C and b) The relation between minimum value of transmittance and different solutions concentration; Maximum value of T%

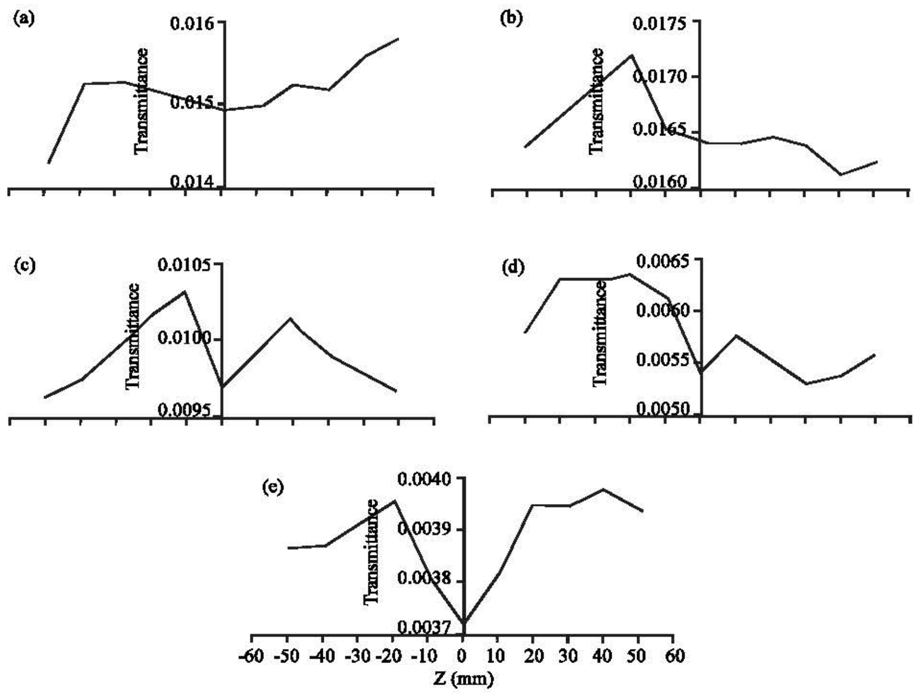

Fig. 3: Open aperture Z-scan of different solutions concentration of RC; a) $7*10^{-6}$; b) $1*10^{-5}$; c) $3*10^{-5}$; d) $5*10^{-5}$ and e) $7*10^{-5}$

Table 1: Some information related to absorption and transmittance of Rhodamine C dissolved in 1-butanol

Concentration (mol/L)	Maximum absorption wavelength (nm)	Absorption intensity	(FWHM) absorption (nm)	Minimum value of T(%)	(FWHM)-T% (nm)
$7*10^{-6}$	545	0.569	18.4125	26.6	36.4735
$1*10^{-5}$	545	0.833	19.4508	14.7	39.8063
$3*10^{-5}$	545	2.232	32.6076	0.6	62.9045
$5*10^{-5}$	545	2.7	40.3811	0.2	73.9139
$7*10^{-5}$	550	2.714	50.7109	0.2	80.3968

Close Z-scan aperture experiments have been used in order to calculate the nonlinear refractive index by using pinhole with diameter (1.5 mm) which placed in front of the detector, Fig. 4 showed the closed aperture curve of Rhodamine C at different concentrations. This indicates that the value of the nonlinear refractive index is negative (self-defocusing), i.e., the transmittance of the top is higher than the bottom transmittance due to the act of the sample as a negative lens that disperses the laser beam.

Table 2: Nonlinear properties of rhodamine C dissolved in 1-butanol

Conc.	Abs. at 532 nm	α (cm^{-1})	L_{eff} (cm)	T (Z)	β (cm/W)	ΔT_{pv}	$\Delta\Phi_0$ $*10^{-6}$	n_2*10^{-11} (cm^2/W)
$7*10^{-6}$	0.4480	20.6348	0.03119	0.01491	6.29572	0.00240	1.2861	2.46179
$1*10^{-5}$	0.6650	30.6299	0.02558	0.01648	7.66162	0.00202	1.0813	2.52287
$3*10^{-5}$	1.8810	86.6388	0.01139	0.00957	17.33306	0.00222	1.18818	6.22791
$5*10^{-5}$	2.6280	121.0457	0.00824	0.00541	24.05507	0.00219	1.17215	8.49096
$7*10^{-5}$	2.6695	122.9572	0.00811	0.00368	24.47234	0.00140	0.74930	11.02490

Table 3: The effect of variation pinhole diameter on nonlinear properties of rhodamine C solution at ($7*10^{-6}$ mol/L)

Pinhole diameter (nm)	ΔTpv	$\Delta\Phi_0$$*10^{-6}$	n_2 (cm^2/W)$*10^{-11}$
1.5	0.00240656	1.2861	2.46179
2	0.00260778	1.39364	2.6676
2.5	0.00323333	1.72794	3.30755

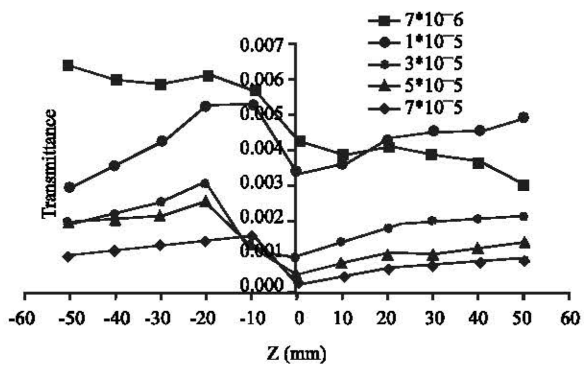

Fig. 4: Close aperture Z-scan of different solutions concentration of RC

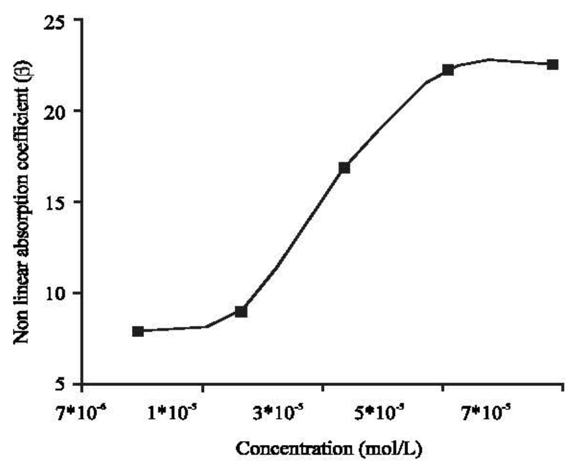

Fig. 5: Relation between nonlinear absorption coefficient and different dye concentration

Table 2 showed the nonlinear refractive index (n_2) value and Fig. 5 and 6 showed the relationship between concentration and nonlinear absorption coefficient and nonlinear refractive index, respectively (Table 3 and Fig. 7).

Effect of changing pinhole diameter: To study the effect of changing the pinhole diameter from which the laser

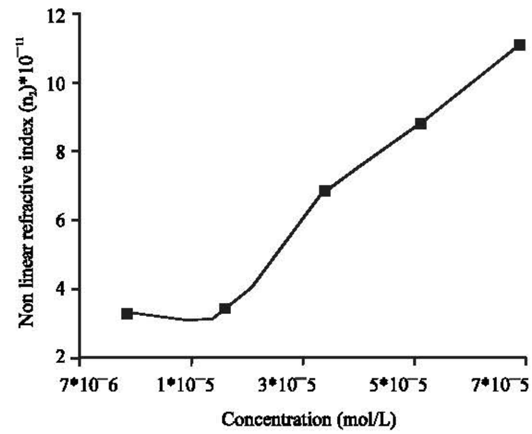

Fig. 6: Relation between nonlinear refractive index and different dye concentration

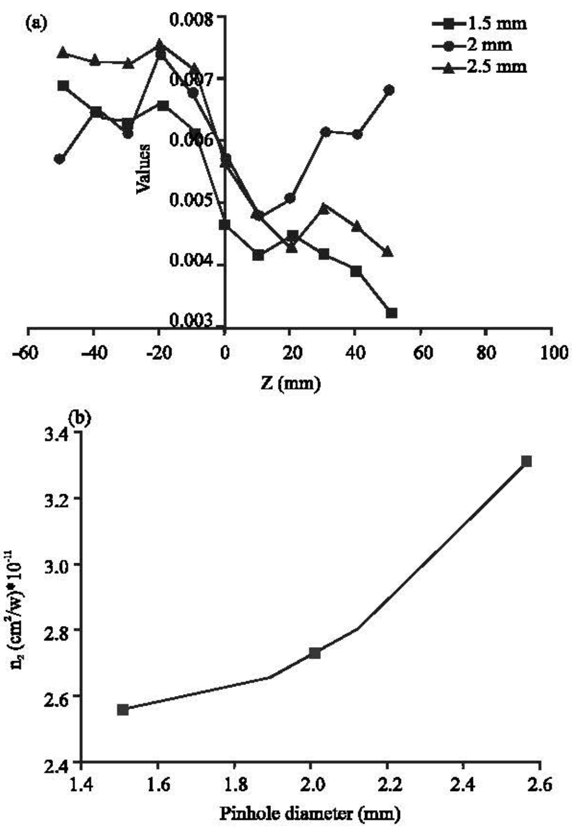

Fig. 7: Close aperture Z-scan of ($7*10^{-6}$ mol/L) Rhodamine C solution in different pinhole diameters

beam is passing, different diameter (1.5, 2 and 2.5 mm) of pinhole was chosen and a random concentration was selected which was ($7*10^{-6}$ mol/L) to study the effect of this variable on its nonlinear properties, Fig. 7 showed the closed aperture curve and Table 3 demonstrates the value of nonlinear refractive index (n_2). The value of (n_2) is increasing due to the increase in the value of nonlinear phase shift ($\Delta\Phi_0$) in addition to increasing in the value of transmittance (ΔT_{pv}) which results from the close aperture Z-scan.

CONCLUSION

From the experimental outcome, we can observed that:

- Absorption intensity is proportional to the dye concentration in the solution
- Nonlinear optical properties are proportional with the increasing of dye concentration
- n_2 increase with increasing pinhole diameter
- β for dye solutions behave like "Two photon absorption"
- n_2 is negative (self-defocusing)

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