Spectral and Third Non-Linear Properties for Mixture Solutions of (R6G,RB, and RC) Dyes

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ABSTRACT

In this research, the spectral characteristics and the nonlinear optical properties for the mixing of Rhodamine dyes (Rh6G, RC, and RB) were determined at different concentrations (1×10^{-5},2×10^{-5},5×10^{-5},7×10^{-5}, and 1×10^{-4} mole/L) at room temperature. The spectral characteristics were studied by recording their absorption and fluorescence spectra. The intensity of absorption increased and fluorescence decreased when increasing concentration which in agreement with Beer – Lambert Law. It was observed that this mixing had a wide spectral range. The quantum efficiency decreased while the radiative life time and the fluorescence life time increased when increasing the concentration. Nonlinear optical properties were measured by using Z-Scan technique, using (CW) continuous Nd: YAG laser with frequency doubled wavelength (532nm) with output power (100 mW). The obtained nonlinear properties results of the mixture (R6G, RC, and RB) showed a negative nonlinear refractive index for concentrations (7×10^{-5}, and 1×10^{-4} mole/L) while the concentrations (1×10^{-5},2×10^{-5}, and 5×10^{-5} mole/L) showed a positive nonlinear refractive index, also this mixing showed two photon absorption behavior for all concentrations. The origin of optical nonlinearity in the dye may be attributed to laser-heating induced nonlinear effect.

Keywords: Xanthenes dye, Rhodamine B, Rhodamine C, Rhodamine 6G, Nonlinear optics, Z-scan technique.

دراسة الخواص الطيفية واللاخطية لمزيج محاليل الصبغات (R6G, RC, and RB)

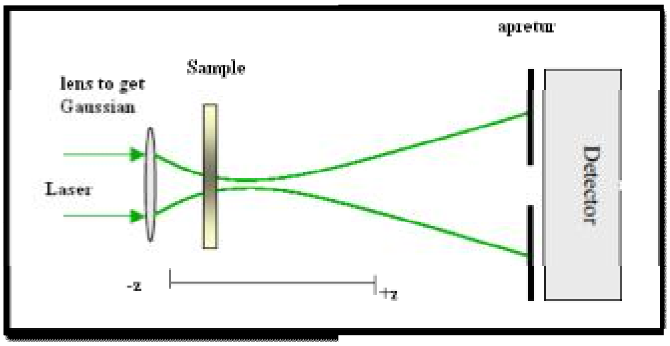
الخلاصة:

في هذا البحث، تم حساب الخصائص الطيفية والخصائص البصرية اللاخطية لخليط أصباغ الرودامين (Rh6G، RC، وRB) لتراكيز مختلفة (1×10^{-5}،2×10^{-5}, 5×10^{-5}, 7×10^{-5}، و 1×10^{-4} مول / لتر) في درجة حرارة الغرفة. تم دراسة الخصائص الطيفية من خلال تسجيل أطياف الامتصاص والفلورة. شدة

الامتصاص زادت وشدة الفلورة انخفضت عندما يزداد التركيز الذي يتفق مع قانون بير-لامبرت. لوحظ أن هذا الخليط له نطاق طيفي واسع. الكفاءة الكمية انخفضت بينما زاد كل من زمن عمر الفلورة وزمن عمر الاشعاع عند زيادة التركيز. تم قياس الخواص البصرية اللاخطية باستخدام تقنية المسح بالبعد الثالث (Z-Scan)، وذلك باستخدام ليزر النيدميوم- ياك المستمر مع التردد المضاعف للطول الموجي (532nm) مع انتاج طاقة (100 ملي واط). أظهرت نتائج الخواص اللاخطية التي تم الحصول عليها من الخليط (R6G، RC، وRB) معامل انكسار لاخطي سالب للتراكيز (7×10^{-5}، و 1×10^{-4}مول / لتر) في حين أن التراكيز (1×10^{-5}، 2×10^{-5}, 5×10^{-5} مول / لتر) أظهرت معامل انكسار لاخطي موجب, كذلك هذا الخليط أظهر سلوك امتصاص فوتونين لجميع التراكيز. ويمكن أن يعزى أصل اللاخطية البصرية في الصبغة لسخونة الليزر بفعل تأثير اللاخطية.

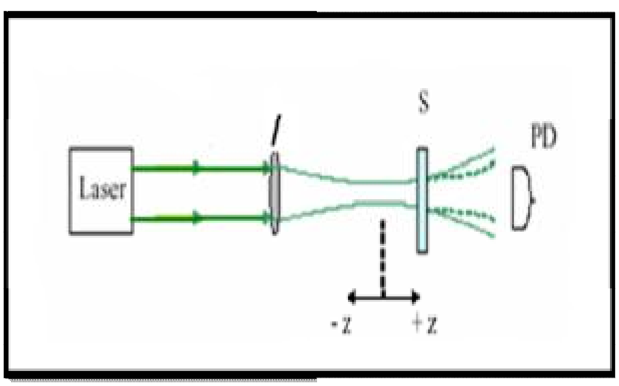
INTRODUCTION

Dye lasers are attractive sources of coherent tunable radiation because of their unique operational flexibility.. The salient features of dye lasers are their tunability, with emission from near ultraviolet to the near infrared. High gain, broad spectral bandwidth enabled pulsed and continuous wave operation[1,2]. Organic dye molecules are widely used in solutions as amplifying media in a tunable lasers[3]. There are large amount of data about laser dyes from many researchers. In (2001) H. M. Mekhlif studied the effect of oxygen on absorption and fluorescence spectrum of two organic dye laser (RB, R6G) in different solvents (methanol, ethanol , and a binary instance sulfur dioxide) and found that the present of oxygen effect on fluorescence spectrum, quantum efficiency, fluorescence life time[4]. In (2011) Ali Hadi Al-Hamdani, Shaima Khyioon, Rafah Abdul Hadi studied the spectral properties of two dyes (RC, and R6G) separately and also for the mixing of these dyes for different concentration, they found that the best concentration was (1×10^{-5} mole/L) which quantum efficiency 96% so this concentration of mixtures can be used to improve solar cell conversion efficiency [5]. In (2013) R. M. Ahmed, and M. Saif studied the spectral characteristics of the prepared samples of dye (RB) doped in the different transparent polymer hosts (PVAc, PMMA, PVAc/PMMA 50/50) by absorption and fluorescence spectroscopy. They concluded that the absorption peaks values as well as the fluorescence intensity were increased by increasing the concentration[6]. Organic material can show very high nonlinear coefficients, because of the large variety of these compounds at high intensities[7]. Many of researchers studied nonlinear properties for many dyes. In (2010) Zainab Fadhil studied the spectral characteristics and nonlinear optical properties of the mixed donor (C480) acceptor (R6G) by using Z-Scan technique, using Q-switched Nd:YAG laser with 1064 nm wavelength. The obtained nonlinear properties results of the mixture C-480/ Rh-6G showed a negative nonlinear refractive index and reverse saturation absorption results show that mixture of laser dyes are effective nonlinear optical materials as compared to individual laser dyes[8]. In (2013) Amal F. Jaffar studied solvents effect on the nonlinear optical properties of oxazine dyes doped films in PMMA at concentration of 10-5 M in three solvents (Trichloroethan, Chloroform ,THF) by using a high sensitive method known as Z-Scan technique The nonlinear refractive index was found to be of the order of 10^{-6} cm^2/W. The magnitude of third order susceptibility was of the order of 10^{-9} cm/watt[9]. Z-scan technique is a simple and sensitive method introduced in 1990 to measure the nonlinear refractive index of optical materials. It is based on a single beam method. It refers to the process of inserting a sample in a focused Gaussian beam and translating it along beam axis through a focal region. The wave front distortion from self-focusing or self-defocusing will cause the kerr nonlinearity. The beam power propagating through a small aperture at far field varies with a sample position. Measuring the output versus position sample allows to determine nonlinearity. There are two methods of z-scan, the closed aperture and open aperture system[10]. A closed-aperture z-scan measures the change in power intensity of a beam, focused by lens L in Fig.(1). Photo-detector PD collects the light that passes through an axially centered aperture A in the far field. The change in on-axis intensity is caused by self-focusing or self-defocusing by the sample S as it travels through the beam waist. A TEM00 Gaussian beam has greatest intensity at the center and will create a change in index of refraction forming a lens in a nonlinear sample as shown in Fig. (1) [11].



Figure(1): The scheme of the closed aperture z-scan

An open-aperture z-scan measures the change in power intensity of beam, focused by lens L in Fig.(2). In the far field at detector PD, which captures the entire beam. The change in power intensity is caused by multi-photon absorption in the sample S as it travels through the beam waist. In the focal plane where the intensity is greatest, the largest nonlinear absorption is observed. At the "tails" of the z-scan signature, where | z | >> zo, the beam intensity is too weak to elicit nonlinear effects. The higher order of multi-photon absorption present in the measurement depends on the wavelength of light and the energy levels of the sample[11]



Figure(2): Open-aperture Z-Scan

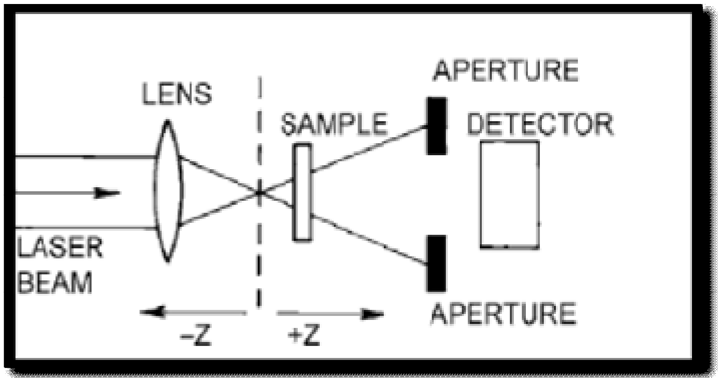
Experimental work

Materials and Instrument

Three well known groups of laser dyes, (R6G, RB, and RC) belong to xanthene family, The molecular formula of (R6G) dye is ($C_{28}H_{31}N_2O_3Cl$), it is observed as dark reddish purple, brown or black crystalline solid. The molecular formula of (RC) is ($C_{28}H_{31}O_3N_2Cl$) and molecular weight (MW= 479.02gm/mole), available as solid crystalline powder. The molecular formula of (RB) is ($C_{28}H_{31}N_2O_3Cl$) and molecular weight (M_W = 479.02gm/mole). These dyes were supplied from (HIMEDIA) India company. These three laser dyes were dissolved in chloroform solvent whose scientific name (Trichloro Methane) with a Chemical formula ($CHCl_3$), molecular weight (Mw=119.38gm/mole) %), refractive index (1.4459), and Productive company Analar company (England) at different concentrations The analytic concentrations of the five solutions examined are 1×10^{-5}, 2×10^{-5},5×10^{-5}, 7×10^{-5} , and 1×10^{-4} mole/L . A 2:2:2 (v/v) solution were mixed to produce five series. The absorption and fluorescence spectra of mixture of Rhodamine dyes were recorded by a UV–VIS–NIR spectrophotometer (SP 8001) supplied from Metertech company which working with wave length (190-1100nm) and SL 147 spectrophotometer supplied from (ELICO limited) Indian company.

Z - Scan experiment

The principle of the Z-scan technique was based on using a Gaussian laser beam in a tight focusing geometry, and moving the sample under investigation along the beam (along the z-axis) through the focal point. The transmittance in the far field was measured and normalized to 1 for linear absorption, and plotted as a function of sample position along the z-axis, with the focus of the laser beam chosen to be at z = 0. The Z-scan experiments were performed using continuous (CW) Nd:YAG laser with frequency doubled wavelength (532 nm) focused by a lens of 12 cm focal length. The laser beam waist w_o at the focus was measured to be 0.0675 mm and the Rayleigh length $Z\boldsymbol{R}$ is 26.9 mm (where $Z\boldsymbol{R}$ is the diffraction length of the laser beam, ($Z\boldsymbol{R}=Kw_o^2/2$), and $K=2\pi/\lambda$¸ is the wave number. The schematic of the experimental setup used is shown in Fig.(3). A 1 mm width quartz cell containing the aqueous solution of mixture of Rhodamine dyes is translated across the focal region along the axial direction, which is the direction of the propagation of the laser beam. The transmission of the beam through an aperture placed in the far field is measured using photo detector operating at wavelength(400-1100) and it is supplied from (Changchun) company and its type is (S121C), with power (1-500mW). For an open aperture Z-scan, a lens replaced the aperture to collect the entire laser beam transmitted through the sample. When the sample is moved from negative Z into focus, initially the beam irradiance is low and negligible nonlinear refraction occurred. Hence the transmittance remained relatively constant. As the sample is brought closer to focus the, beam irradiance increased leading to self-lensing in the sample. A negative self-lensing prior to focus collimated the beam and caused beam narrowing at the aperture, which resulted in an increase in transmittance. As the sample is moved away from the focus i.e., towards positive Z the beam divergence caused a decrease in transmittance at the aperture. A pre-focal transmittance maximum (peak) followed by a post-focal transmittance minimum (valley) is the z scan nature of negative nonlinearity. The opposite effect is the nature of positive nonlinearity. The sensitivity to nonlinear refraction is entirely due to the aperture, and the removal of aperture completely eliminates the effect. The third order nonlinear refraction index of the sample (Dye solution) was evaluated from the Z-scan data. The Z-scan is performed for different polarity of organic solvents at different concentrations of the dye solution.



Figure(3): Z-Scan system .

Quantum efficiency

It is the ratio of the number of fluorescence photons emitted by a system of molecules in dilute solution to the number of molecules excited into S_1 (the number of absorbed photons). Quantum efficiency is given by the following equation:

$$qfm = \frac{Number\ of\ quanta\ emitted}{Number\ of\ quanta\ absorbed} \quad (1)$$

The Radiative life time (τ_{fm})

The raditive life time (τ_{fm}) is given by using the following relation:

$$\frac{1}{t_{FM}} = 2.88 * 10^{-9} n^2 (n'^2) \int e('n) dn' \qquad ... (2)$$

where

n:refractive index of a medium

ύ: wave number at the maximum absorption

∫ε(ύ)d ύ:the area under the absorption spectrum curve as a function of the wave number[12].

Nonlinear refractive index

Nonlinear refractive index was measured by close aperture z-scan technique by difference between peak and valley for transmittance according to equation:-

$$\Delta T\ p\text{-}v = 0.406\,(1-S)^{0.27}|\Delta\phi o \qquad ... (3)$$

$\Delta Tp\text{-}v = T_P - T_V$ the change between peak and valley. S is the aperture transmittance is given by[10]:

$S = 1 - \exp(-2 r^2_a/\omega^2_a)$ … (4)

r_a : representing the aperture radius, ω_a representing the radius of the laser spot before the aperture[13]. In this experiment the value of S= 1.12×10^{-4} then $(1\text{-}1.12\times10^{-4})^{0.27}$= 0.999≈ 1) so ΔT p-v become:

ΔT p-v = 0.406 |ΔΦo|

So the nonlinear refractive index calculated from this equation:

$n_2 = \Delta\Phi_o / I_o L_{eff}\ k$ … (5)

$\Delta\ \Phi_0$:- the change in nonlinear phase, also L_{eff} represent effective length for sample (L_{eff} = $(1\text{-}\exp^{-\alpha0L})/\ \alpha_O$) ,and k wave facter equal($k = 2\pi /\lambda$)[10], and I_o represents the intensity of beam in focus equal ($I_O = 2P/\ \pi W_{or}^2$)[14]. and α_O represents linear absorption equal ($\alpha_0 = 1/t \ln 1/T$)[11].

Nonlinear absorption coefficient

Nonlinear absorption coefficient was measured by open aperture z-scan technique from transmittance curves according to equation[11,14] .

$T(z) = 1\text{-}\ q_0 / 2\sqrt{2}$ **...** (6)

$q_0 = \beta\ \text{Leff}\ I_0 [1+z]$. … (7)

The real and imaginary parts of the third-order nonlinear optical susceptibility $\chi^{(3)}$ are defined as :

$\text{Re}\ \chi^{(3)}\ (\text{esu}) = 10^{-4}\ \varepsilon_0\ c^2\ n_o^2/\ \pi$ … (8)

$\text{Im}\ \chi^{(3)}\ (\text{esu}) = 10^{-2}\ \varepsilon_0\ c^2\ n_o^2\ \lambda\ \beta /4\pi^2$ …(9)

where

ε_0 is the vacuum permittivity and *c* is the light velocity in a vacuum. The absolute value of $\chi^{(3)}$ is calculated from[15].

$\chi^{(3)}=\{[\text{Re}\chi^{(3)}]^2+[\ \text{Im}\chi^{(3)}]^2\}^{1/2}$ … (10)

Results and Discussion

Spectral result

The spectra of absorption and fluorescence was studied for mixture of dyes (R6G, RC, and RB) dissolved in chloroform to concentrations ($1\text{x}10^{-5}$, $2\text{x}10^{-5}$, $5\text{x}10^{-5}$, $7\text{x}10^{-5}$, and $1\text{x}10^{-4}$ mole / L) as shown in fig. (5) and (6) and found that the increasing of concentration shifted the absorption spectrum towards shorter wavelengths (Blue Shift) and shifted the fluorescence spectrum towards longer wavelengths (Red Shift) because of non-radiative process (Internal conversion, Inter system crossing. Fig.(4) indicates the absorption and fluorescence spectra of the chloroform did not absorb at the same rang of dye solution. Stock's shift increased as the dye concentrations was increased for all dyes. Tables (1) shows the important parameters, the raditave τ_{fm} (nsec) and fluorescence life time τ_f (nsec) and the quantum efficiency Φ_{fm} of the mixture. The quantum efficiency decreased as the dye concentrations was increased for dyes because of decrease the probability of non-radiative transition (I.S.C and I.C.).The raditave $\boldsymbol{\tau_{fm}}$(nsec) and fluorescence life time τ_f (nsec) increased as the dye concentrations was increased for dyes. These results in agreement with results of [5, 16, and 17]. These results indicate that mixing the dyes may improve the characteristics properties of the RC,R6G,RB dye and can be used in different applications (luminous solar concentrators and laser dye medium).

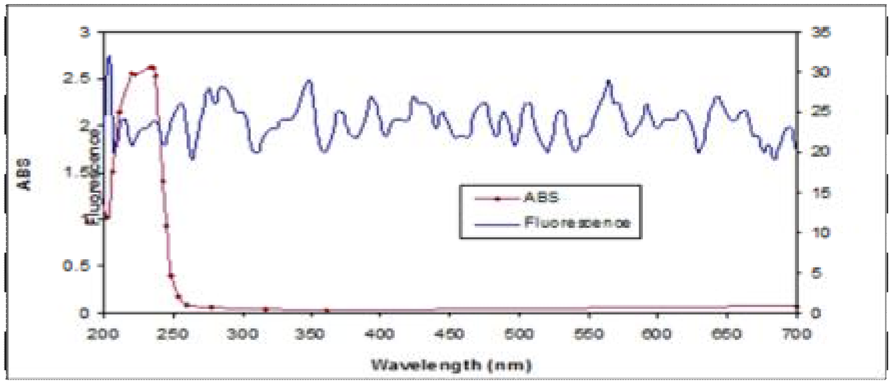


Figure (4): The absorption and Fluorescence spectrum of chloroform

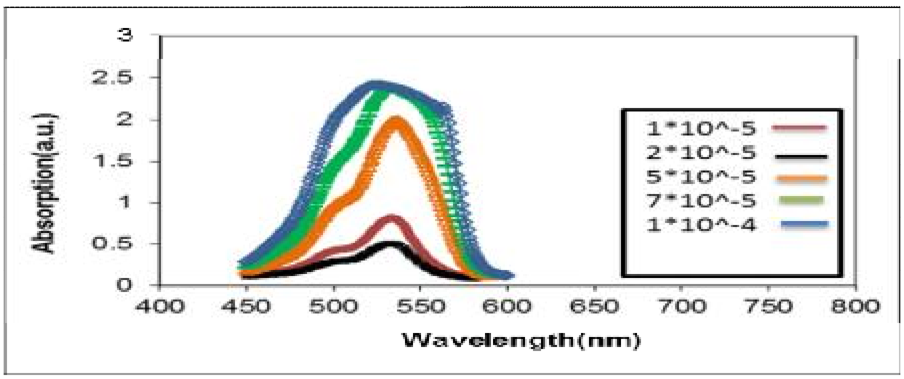


Figure (5): absorption spectra for mixture (RC+RB+R6G) at different concentration

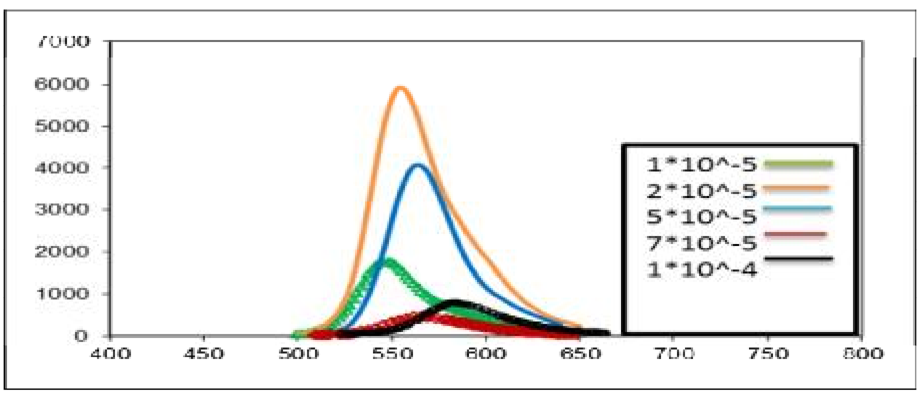


Figure (6): Fluorescence spectra for mixture (RC+RB+R6G) at different concentration.

Table (1) Spectral parameter for mixture of (RC+RB+R6G)

C mole/L	$\lambda_{abs.}$ nm	$\acute{\upsilon}_{abs.}$ cm^{-1}	λ_{flu} nm	$\acute{\upsilon}_{flu.}$ cm^{-1}	Stock Shift	Quantum efficiency % (q_{FM})	Kfm sec^{-1}	τ_{FM} Nsec	τ_F nsec
1×10^{-5}	533	18761.73	549	18214.936	16	90.35	6960071.214	1.44	1.29
2×10^{-5}	533	18761.73	554	18050.541	21	85	5764663.096	1.73	1.47
5×10^{-5}	536	18656.72	564	17730.496	28	78	5026850,524	1.989	1.55
7×10^{-5}	535	18691.59	570	17543.859	35	79	4770071.398	2.09	1.65
1×10^{-4}	526	19011.41	584	17123.287	58	65.16	4315932.9	2.13	1.5

Z - Scans results

Experiment is performed for different concentrations of the dye solution, A typical closed aperture Z-scan curve of the dye solution at concentrations ($1x10^{-5}$, $2x10^{-5}$, $5x10^{-5}$, $7x10^{-5}$, and $1x10^{-4}$ mole/L) exhibiting the normalized transmittance is shown in the Fig. (7) at incident intensity Io= 1.39 KW/cm^2.The curves are characterized by a pre-focal peak followed by a post-focal valley for some concentration ($7x10^{-5}$,$1x10^{-4}$ mole/L) and revers for other concentrations (1×10^{-5},2×10^{-5} and 5×10^{-5} mole/L), which implies that the nonlinear refractive index is negative (n_2<0) for concentration($7x10^{-5}$,$1x10^{-4}$ mole /L) and positive for concentration(1×10^{-5},2 $\times10^{-5}$ and 5×10^{-5} mole/L). The transmittance profile at open-aperture (OA) z-scan experiment shows (Two Photon Absorption) for concentrations ($1x10^{-5}$,$2x10^{-5}$, $5x10^{-5}$,$7x10^{-5}$, and$1x10^{-4}$ mole /L) as in Fig.(8). To avoid any discrepancy caused by deviations from an ideal Gaussian profile, all the measurements were taken with the experimental configuration kept identical for all the concentrations of the dye. Investigation has been carried out for the dependence of nonlinear refractive index on concentration. The nonlinear refractive index n_2 increased with increasing concentration as in Fig.(9). This may be attributed to the fact that the number of the dye molecules increases when the concentration increases and more particles get thermally agitated resulting in an enhanced effect . The nonlinear refractive index (n_2) and nonlinear absorption coefficient (β) values of the dye solution are given in Table (2) and the corresponding $\chi^{(3)}$ values calculated from data of nonlinear refractive index and nonlinear absorption coefficient. The linear dependence of nonlinear absorption coefficient β on the concentrations of mixture of (R6G, RC, and RB) as shown in Fig. (9). The third order nonlinear susceptibility $\chi^{(3)}$ increases with increasing concentration as in Fig. (10).All results of n_2 , β and $\chi^{(3)}$ show a direct relation with concentration.

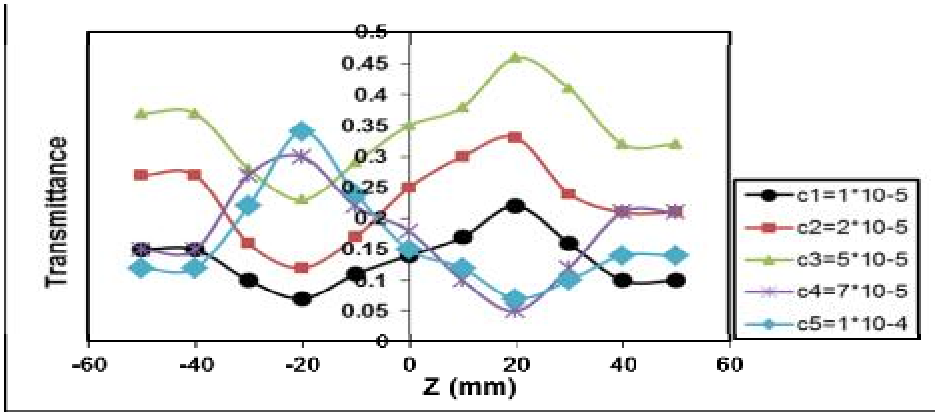
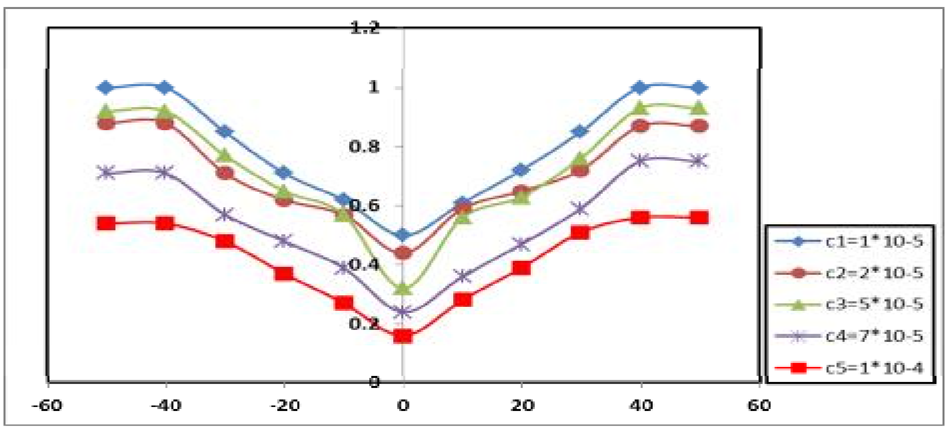
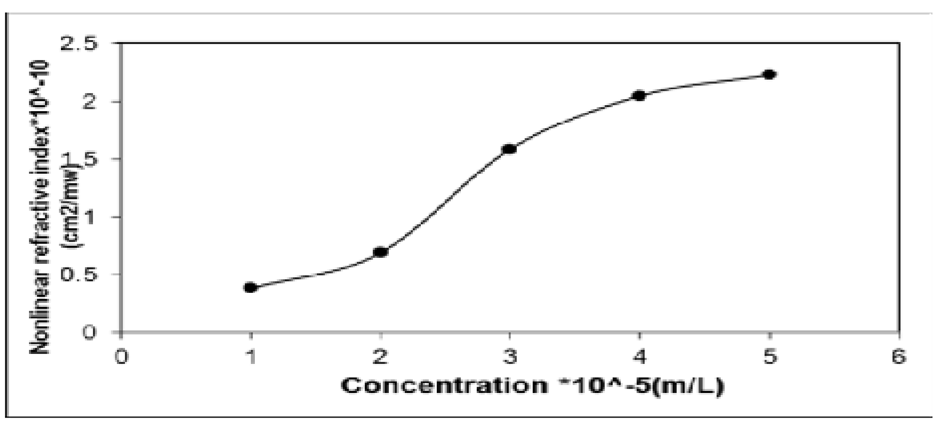


Figure (7): Closed aperture Z scan data for different concentration of aqueous solution mixture of Rhodamine dyes at I_0 = 636.6 W/cm^2 and (S=1.12 ×10^{-4})



Figure(8): Open aperture Z scan data for different concentration of aqueous solution mixture of Rhodamine dyes at I_0 = 636.6 W/cm2 and (S=1)



Figure(9): Relationship between the nonlinear refractive index (n_2) and concentration

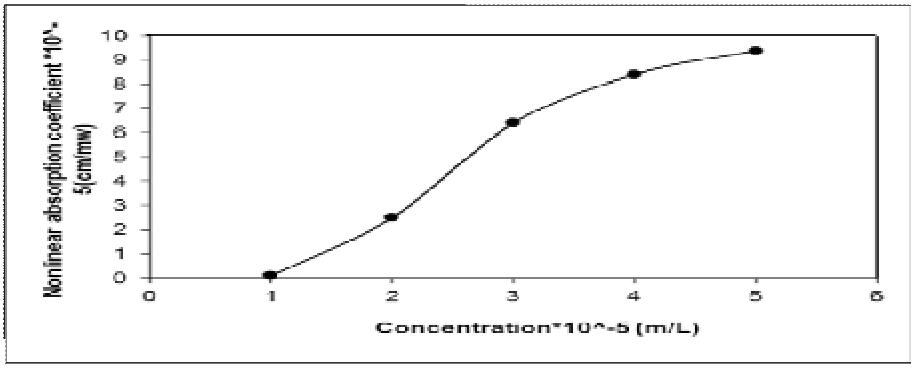


Figure (10): Relationship between the nonlinear absorption coefficient (β) and concentration

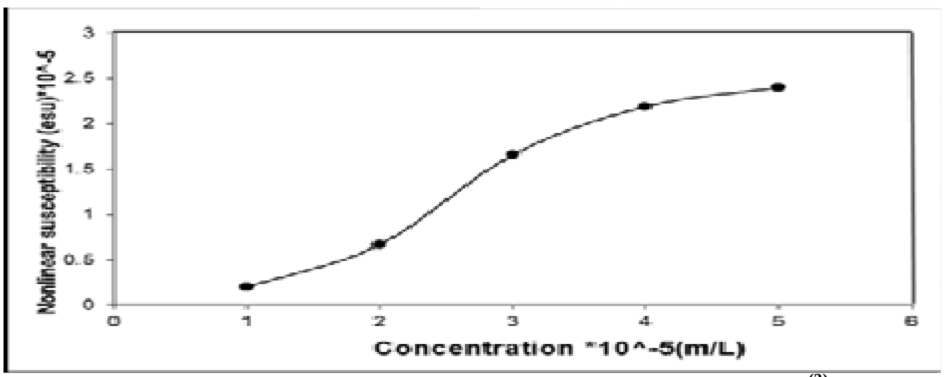


Figure (11): Relationship between the nonlinear susceptibility ($\chi^{(3)}$) and concentration.

Table (2) Nonlinear parameters for mixture of (RC, RB, and R6G)

C mole/litter	T%	α cm^{-1}	n_o	L_{eff} cm	ΔT_{pv}	$\Delta\Phi_0$	n_2 cm^2/mw	T(z)	β cm/mw	$\chi^{(3)}$ esu
1×10^{-5}	30.01	12	1.4458	0.0582	0.15	0.369	3.8×10^{-11}	0.50	1.8×10^{-5}	4.5×10^{-6}
2×10^{-5}	15.33	18.7	1.4459	0.0452	0.21	0.517	6.9×10^{-11}	0.44	2.5×10^{-5}	6.7×10^{-6}
5×10^{-5}	1.06	46	1.4460	0.0215	0.23	0.566	1.5×10^{-10}	0.32	6.4×10^{-5}	1.6×10^{-5}
7×10^{-5}	0.41	54.9	1.4465	0.0181	0.25	0.615	2×10^{-10}	0.24	8.4×10^{-5}	2.1×10^{-5}
1×10^{-4}	0.39	55.4	1.4467	0.0179	0.27	0.66	2.2×10^{-10}	0.16	9.4×10^{-5}	2.4×10^{-5}

CONCLUSIONS

The spectral properties of mixture solution of Rodamine dyes (B,C,6G) soluble in chloroform were studied. The results indicate that the best concentration was (1×10^{-5} mole/L) which quantum efficiency equal (90.35%) We have also measured the nonlinear refraction index coefficient n_2, the nonlinear absorption coefficient β, and susceptibility $\chi^{(3)}$ for mixture solution by using *Z*-Scan technique. *Z*-Scan measurements indicated that the dye exhibited negative nonlinear refractive index for some concentration ($7x10^{-5}$,$1x10^{-4}$ mole/L) and exhibit positive nonlinear refractive index for concentration (1×10^{-5},2×10^{-5}, and 5×10^{-5} mole/L). We also observed that these dyes exhibited (Two Photon Absorption) behavior for concentrations ($1x10^{-5}$,$2x10^{-5}$, $5x10^{-5}$,$7x10^{-5}$,and$1x10^{-4}$ mole/L) , All of these experimental results show that mixture of these dyes dye is a promising material for application in nonlinear optical devices.

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