Study the Linear and Nonlinear Optical Properties for Laser Dye Rhodamine B

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Abstract. In this study different concentrations ($10^{-2}$, $10^{-3}$, $10^{-4}$) mol/l were prepared for Rhodamine B dye in solvent water at room temperature, then the optical linear properties for example transmission and absorption spectrum were calculated by employ UV-visible Spectrophotometer to find the linear index of refraction, the linear coefficient of absorption, the coefficient of extinction, and reflectivity, the results showed an increase in the coefficient of extinction and index of refraction values with the increase in the concentrations. The properties of nonlinear optical of the samples for example non-linear index of refraction, and non-linear coefficient of absorbance were measured by using a new technical method called the $Z$-Scan technique, in two parts; The first one was a closed aperture placed ahead of the detector to calculate the index of nonlinear refractive. While the second part; the aperture ahead the detector was taken away (open manhole) to calculate the coefficient of nonlinear absorption. The two cases were performed using diode laser of a continuous wave (CW) with 650 nm wavelength and input power of 50 mW. The result shows that the samples exhibit negative refractive index ($-n_2$) self-defocusing for all concentrations and two-photon absorption for open aperture $Z$-Scan so that the laser dye at this study can be applied in nonlinear optics application and it is useful in the optical limiter applications.

1. Introduction
Laser dyes are Hydrocarbons unsaturated compounds where the advantage of its containing to double or triple bonds is conjugated with chains of carbon atoms. This system has the advantage of light absorbed in the area(200-700nm) and the transmission spectrum of absorption in the visible region. It can be used as a laser medium in a dye lá.ser. The dyelaser has an important role in the history of the study and development of optical spectroscopy [1]. The dye laser providing much higher spectral brightness compare to the conventional lamp sources. The gain media of solid state laser have a large tuning range gave a limited gain in the infrared region of the spectrum, till that time the dye laser will keep on being the workhorse for experiments requiring tunability in the visible. Consequently, much work has gone into designing and engineering dye laser sources[2].

The scheme for dye laser emission is shown in Figure 1. Dye molecules were excited into the excited states by the light source as a pump. this made all of the molecules dropping into the lowest excited state. There were a lot of atoms in the primary vibrational dimension of the ground electronic state contrasted with the most reduced energized state, however a populace reversal exists as for higher vibrational dimensions of the ground state. Invigorated discharge inside the laser cavity at that point results, with the change of energized electronic states into vibrationally energized ground state
atoms. Since numerous last vibrationally energized dimensions are accessible, the laser outflow can be tuned over thin ranges[3].

![Figure 1](image1.png)

**Figure 1.** (a) The scheme for dye laser action. (b) Energy loss with laser action for dye laser [3].

2. Materials and Methods:

2.1. Characteristics of Rhodamine B

The vast majority of the dye lasers are from this class – Fluorescein and Rhodamine B were the two extensively utilized laser dyes. The electron conveyance in the xanthene’s dyes can be portrayed by the accompanying two indistinguishable structures, (22) and (23)[3].

![Figure 2](image2.png)

**Figure 2.** The π-electron distribution in the chromophore in the xanthene dyes

Rhodamine B dye belongs to the xanthine family. It is one of the most commonly used in various spectroscopic Studies [4]. According to their spectral properties, rhodamines amino kind of xanthene are broadly used in technology and science for lasing, and in thermochemistry as fluorescence labels and probes for investigating different objects (biological and non-biological). Rhodamine B, a red xanthene dye, had been widely used in dye lasers [5] and as a fluorescent label in biological staining[6].

![Figure 3](image3.png)

**Figure 3.** Rhodamine B dye [7]

The properties of this dye is presented show in table (1).
2.2. Linear Optical Properties

2.2.1 Absorption: is the ratio of the absorbed intensity by the matter to the original incident intensity, as [8].
\[ A = \frac{I_A}{I_0} \]……………………………………(1)

2.2.2. The Transmittance: is definite as the proportion of transmitted beam intensity from the matter to incident intensity as [8]:
\[ T = \frac{I_T}{I_0} \]………………………………………(2)

2.2.3. The reflectance: is defined as the ratio of reflected light intensity from the matter to incident intensity, as [8]:
\[ R = \frac{I_R}{I_0} \]………………………………………(3)

The reflectance value (R) was calculated by using the equation (4) below;
\[ R = 1 - (A + T) \]………………………………………(4)

Where T is the Transmittance.

2.2.4. The linear coefficient of absorption \( \alpha_o \): is definite as the proportion of the decrease in the energy of the incident beam to the distance towards the spread of the wave inside the matter, the absorption coefficient depends on the energy of the incident photon and the length of the absorbent centre path as [9]:
\[ \alpha_o = 2.303 \times \frac{A}{t} \]………………………………………(5)

Where A: the Absorbance, t: is the sample thickness.

2.2.5. Extinction coefficient: is the quantity of energy that absorbed by valance band electrons or is the quantity of attenuation in incident beam energy [10]. The extinction coefficient dependents on the coefficient of absorption for each matter and the magnitude of the wavelength for incident beams and can be calculated from:
\[ K = \alpha_o \lambda / 4\pi \]………………………………………(6)

where (\( \lambda \)) is the incident radiation wavelength.

2.2.6. The index of refraction: is the proportion between the speed of light in the vacuum to the speed of light inside the matter. Thus, it is significant to calculate the optical constants of the sample and it is calculated from [11]:
\[ n = \frac{(4R-K^2)_{0.5}}{(R-1)^2} - \frac{R+1}{R-1} \]…………………………………………(7)

Where R is the reflection and K is the coefficient of extinction.

2.2.7. Optical Energy Gap: the estimation of optical energy gap is important to build up the electronic band structure of the laser dye. The energy of band gap of the samples were determined from the absorption spectrum using the relation between the coefficient of absorption (\( \alpha_o \)) and the of incident light energy \( hv \) [12].
\[ \alpha_o hv = P (h v - Eg)^r \]………………………………………(8)
Where $h\nu$ is the energy of photon, $P$ is continual, and $r$ represents transition type ($r=1/2$ for direct transition, $r=2$ for indirect transition) and by drawing a graph between $(\alpha h\nu)^{0.5}$ and photon energy $(h\nu)$ for direct allowed transitions, and draws the straight part to cut axis point (X) of photon energy at the point where $((\alpha h\nu)^{0.5} = 0)$, which represents the value of the optical energy gap.

2.2.8. The complex dielectric constant ($\varepsilon$): The dielectric function of a thin film is a complex quantity which consists of both the imaginary and real parts. It is a major characteristic property of the material [13]:

\[ \varepsilon = (\varepsilon_1 - i\varepsilon_2) \]

So, the imaginary and real parts of the dielectric constant are connected to the $n$ and $K$ standards from which arise [14]:

\[ \varepsilon_1 = n^2 - K^2 \]

\[ \varepsilon_2 = 2nK \]

2.3. Z-Scan Technique

In 1989 M. Sheik Baha[15], established a calculated method to calculate the nonlinear refraction of thin samples. This is called the Ž-Scan technique[12]. The essential geometry is shown in Fig.4[16].

The Ž-Scan technique include that the solution is scanned ahead the Z direction concluded the beam waist of a focused Gaussian laser beam in a fitted focusing configuration, as shown in Figure 4. As the solution methods focus the spot size decreases, increasing the irradiance on the solutions and the persuaded non-linear effects[17].

There were two parts of the Z-scan: Closed Aperture and Open Aperture.

2.3.1. Closed aperture Z-Scale

Closed Z-scan aperture was an instance of self-refraction phenomenon or self-phase inflection in space. Without absorption of nonlinearity, a known valley and peak were showed.

By observing the transmittance change through an aperture placed away from the detector, to be able to calculate the nonlinear index of refraction. The solution was moved in a step of 1 cm during the scan[18].

The relationship between the normalized transmission $T(z)$ and $z$ position was taken by moving the sample along the axis of the beam direction (z-direction) to the focal point, We distinguish a measurable quantity $\Delta T_{p-v}$ as the difference between the normalized peak and valley transmittance: $\Delta T_{p-v}$ is linearly dependent on the temporally averaged induced phase distortion. The variety of this magnitude as a function of $\Delta \Phi_o$ is given by [19]:

\[ \Delta T_{p-v} = 0.406 \Delta \Phi_o \]
Where $\Delta T_{p-v} = T_p - T_v$  

(13)  

$\Delta T_{p-v}$: the difference in transmittance between the peak and valley.

The non-linear refractive index can be obtained from the formula:

$n_2 = \Delta \Phi_0 / I_0 L_{eff} k$  

(14)

$k = 2\pi/\lambda$  

(15)

$k$: wavelength of laser beam.

$I_0 = 2p/\pi W_0^2$  

(16)

$I_0$: is intensity of the laser beam at the focus ($Z=0$). 

$P$: power of laser beam $w_0$; the beam radius at the focal point.

$L_{eff} = (1 - \exp^{-a_0 t})/a_0$  

(17)

$L_{eff}$: effective thickness of the sample, $t$: is the thickness of the sample.

2.3.2. Open Aperture Z-Scan:

The open aperture Z scan method, was used to calculate the nonlinear coefficient of absorbanance ($\beta$) (Figure 6). For instance, in the event that non-linear ingestion like two-photon assimilation (TPA) is available, it is showed in the estimations as a transmission least at the point of focal point. Then again, if the solutions is a saturable absorber, transmission increments with an expansion in episode force and results in a transmission most extreme at the central district. It has been demonstrated that the model initially created by Bahae et. al for unadulterated TPA can likewise be connected to energized state absorption[20].

![Figure 6. Z-scan: open aperture geometry](image)

In reality, we can be counted the coefficients of the nonlinear absorption from the normalized transmission curves, from the above equation[16].

$$\beta = \frac{2\sqrt{2}}{I_0 L_{eff}} \Delta T$$  

(18)

Where $\Delta T$ is the valley value at the aperture when it is open for Z-scan bend in the graph. The results of $\beta$ could be positive to the absorption saturable and negative for absorption of two-photon.[21]

The result of the real and imaginary parts of the third order optical nonlinear susceptibility $\chi^{(3)}$ are calculated from experimental determination of $n_2$ and $\beta$ according to these equations[22]:

$$\text{Re } \chi^{(3)} = 10^{-4} \frac{6\alpha_0^2 n_i^3}{\pi} \left( \frac{cm^2}{W} \right)$$  

(19)

$$\text{Im } \chi^{(3)} = 10^{-2} \frac{6\alpha_0^2 n_i^3 k \beta}{4\pi^2} \left( \frac{cm^2}{W} \right)$$  

(20)

Where $c$: is the speed of the light, $n_i$: the linear index of refractive The absolute value is counted from the below equation:

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

(21)

3. Experimental part:

3.1 Preparation of the Rhodamine B concentrations:

Three concentration ($10^2, 10^3$ and $10^4$)mol./l (Figure 7), were prepared by dissolving 0.0478 g of RB powder in 10 ml of deionized water. The powder is weighting using an electronic balance Germany origin. By using the following equation:

$$W = \frac{M_w \times V \times C}{1000}$$  

(22)

Where:

$W$: weight of the dissolved dye (gm), $M_w$: molecular weight of the dye (gm/mol.),
V: the volume of the solvent water (ml), C: the dye concentration (mol/l)
The mixing was stirred by using magnetic stirrer at room temperature to get a homogeneous solution.

Figure 7. Three concentration of rhodamine B

3.2. UV-Visible Spectrophotometer:
Spectrophotometer UV-Visible (UV-1800) was provide from SHIMADZU used to measure the absorption and transmission spectra of the Rhodamine B dye, over wavelength range 190-1100 nm.

3.3. Z-Scan technique:
The Z-Scan technique (figure 8) was used to measure the nonlinear optical properties of Rhodamine B solutions in three concentration by using diode laser with wavelength of (650nm) and maximum output power of (50mW), and lens which have focal length (30cm). The sample put in quartz cell was scanned using transition system along direction z-axes through the focusing area. Z-Scan technical consist of two steps the first one called the close aperture z-scan measured the nonlinear refractive index of the sample by the study of the transmission curve and the second one called the open aperture Z-Scan measured the nonlinear absorption coefficient \( \beta_2 \).

Figure 8. Z-scan technique geometry

4. Results and Discussion

4.1. The optical properties linearity of Rhodamine B dye
The linear optical properties for the liquid dye solution (Rhodamine B) includes, transmition, absorbance coefficient, coefficient of extinction, reflectance and index of refraction that are explained in table (2).

Table 2: The result of linear optical properties for Rhodamine B  
| Con. (mol/l) | \( \lambda \) (nm) | Abs. | T% | \( \alpha_o \) (cm\(^{-1}\)×10\(^{-8}\)) | K ×10\(^{-6}\) | R | n |
|--------------|-------------------|------|----|-------------------------------|-----------------|---|---|
| 1×10\(^{-2}\) | 650               | 0.179| 0.63808 | 53.3                      | 0.533116       | 0.31717 | 3.578529 |
| 1×10\(^{-3}\) | 650               | 0.007| 0.91658 | 2.08                      | 0.020848       | 0.08175 | 1.800805 |
| 1×10\(^{-4}\) | 650               | 0.002| 0.92078 | 0.596                     | 0.005959       | 0.07872 | 1.779982 |
4.1.1. Spectra of absorption

The spectra of absorption were measured from the UV-Visible spectrophotometer for three different concentrations \((1 \times 10^{-2}, 1 \times 10^{-3} \text{ and } 1 \times 10^{-4})\) mol./l of rhodamine B (RB), from the curve of the absorbance with wavelength Figure (9) showed that the absorption curve of RB for different concentrations is steady constant at the wavelength from 540 nm to 635 nm with the decrease in the concentration then rapidly the curve for different concentrations was decrease with increase of the wavelength.

![Figure 9: The Spectra of absorption of Rhodamine B dye solution](image)

4.1.2. Transmittance

Transmittance spectra studied in wavelength range \((540-690)\) nm for rhodamine B at different concentrations. The transmittance spectra depends on many factors such as quantity of energy levels, which connects in chemical structure. Figure (10), showed the transmittance spectra for rhodamine B at different concentrations as function for wavelengths. The transmittance spectra is reversed in behaviour to the absorbance spectra. It illustrates that the transmittance has a decreased with value at short wavelength then increased the transmittance with the increasing of the wavelength.

![Figure 10: Transmittance spectrum of Rhodamine B dye solution](image)

4.1.3. Reflection Spectrum

Reflection spectrum is calculated from absorption and transmission spectrum according to eq.(4). Figure (11) explain the different behaviour of reflection spectrum as a function of photon energy for different concentrations as it is seems the relation of Reflectance and photon energy first it is steady then suddenly increased with increasing photon energy to reach the peak at \((1.9, 2.02 \text{ and } 3.6)\) eV for \((10^{-3}, 10^{-4} \text{ and } 10^{-5})\) mol./l respectively, and then drops quickly.
4.1.4. **Extinction coefficient**

The extinction coefficient is calculated from the eq.(6). Fig.(12) illustrates the extinction coefficient of Rhodamine B dye solution at different concentrations as a function of wave length. The curve is seems to be stable decrease in the concentration is seen to increase gradually as the photon energy increases, reaches a maximum, thereafter, decreases with photon energy. The maximum value of extinction coefficient at (1.88, 1.8 and 0.57), obtained at the photon energy of (1.93, 2.21 and 1.93 eV).

4.1.5. **The linear absorption coefficient \( \alpha \)**

The linear absorption coefficient was calculated for the three concentration B\textsubscript{y} using equation (5) the results explained that the absorption coefficient of Rhodamine B, first it is constant then it increased rapidly with the increase of photon energy, as the concentration decrease. It reaches highest value figure (13), for absorption coefficient with [9.507×10\textsuperscript{-6}, 8.902×10\textsuperscript{-6} and 8.184×10\textsuperscript{-6} ](cm\textsuperscript{-1}) at photon energy [2, 2.08 and 2.15] eV respectively, and thereafter, the result became stable with photon energy.

![Figure 1](image1.png)

**Figure 11:** Reflection spectrum of Rhodamine B dye solution

![Figure 2](image2.png)

**Figure 12:** The extinction coefficient for different concentration of Rhodamine B dye solution

![Figure 3](image3.png)

**Figure 13:** The linear absorption coefficient for different concentration of Rhodamine B dye solution
4.1.6. The refractive index
The index of refraction was useful for optical constants and calculated from the eq. (7). Fig. (14) shows the behaviour of refractive index of Rhodamine B dye solution at different concentrations. Decreased concentrations of Rhodamine B dye solution led to increasing the refractive index as the photon energy increase, and thereafter, decreases with the increasing of photon energy.

![Figure 14: Refractive index for different concentration of Rhodamine B dye solution](image)

4.1.7. Optical Energy Gap
The optical energy gap is calculated from the eq. (8). Fig. (15) illustrate the case of the Rhodamine B for three concentrations, by drawing a relationship between \((\alpha h\nu)^{0.5}\) and incident photon energy \((h\nu)\), and draws the straight part to cut axis point (X) of photon energy at the point where \((\alpha h\nu)^{0.5} = 0\), which represents the value of the optical energy gap (1.875, 1.95 and 2.048) eV at the concentrations under the study.

![Figure 15: The variation of the \((\alpha h\nu)^{0.5}\) with the incident photon energy.](image)

4.1.8. The dielectric constant \((\epsilon)\)
The real \(\epsilon_1\) and imaginary \(\epsilon_2\) part of the dielectric constant were determined from equations (10) and (11) respectively. These values depend on the wavelength were shown in Figures (16 a.) and (16 b.) respectively. The \(\epsilon_1\) values are more than that of \(\epsilon_2\) values. It is explain that the \(\epsilon_1\) and \(\epsilon_2\) values increase with increasing of the wavelength and then decreasing with it, for all concentrations.

![Figure 16: The differences of a. Real and b. Imaginary dielectric constant with the photon energy](image)
Table (3) explain the results of the non-linear optical susceptibility from its real and imaginary parts, for three concentrations of Rhodamine B were calculated by using equations (19), (20) and (21) at wavelength 650 nm and output power 50 mW.

Table 3: The outcomes of the real and imaginary parts of the third order non-linear optical susceptibility $\chi^{(3)}$

| Con. (mol./l) | Incident Intensity (mW/cm$^2$) | Re. $\chi^{(3)}$ (cm$^3$/W) | Im. $\chi^{(3)}$ (cm$^3$/W) | $\chi^{(3)}$ |
|---------------|-------------------------------|-----------------------------|-----------------------------|-------------|
| $1\times10^{-2}$ | 14154282 | $3.294\times10^{-6}$ | $1.39\times10^{-16}$ | $3.294\times10^{-6}$ |
| $1\times10^{-3}$ | 14154282 | $8.344\times10^{-7}$ | $2.66\times10^{-17}$ | $8.344\times10^{-7}$ |
| $1\times10^{-4}$ | 14154282 | $6.089\times10^{-7}$ | $1.72\times10^{-17}$ | $6.089\times10^{-7}$ |

4.2. The non-linear optical properties of rhodamine B dye

The Z-Scan results were calculated by 650 nm diode laser (CW), (max output is 50 mW, Ac: 220-240 volt, Frequency: 50-60 Hz 250mA. beam diameter: 1.5 mm, which is focused by 30 cm focal length lens. The laser beam is 0.015 mm. The result of nonlinear optical properties was measured by this technique in two parts close and open aperture Z-scan as shown in table (4).

Table 4: The results of nonlinear optical properties for Rhodamine B at different concentration

| Con. (mol./l) | $\lambda$ (nm) | $\Delta T_{\text{p-v}}$ | $I_o$ (mW/cm$^2$) | $\Delta \Phi_o$ (Rad) | $n_2 \times 10^{-12}$ (cm$^2$/mW) | $T_{\text{max}}$ | $\beta$ (cm/mw) $\times 10^3$ |
|---------------|------------|-----------------|----------------|-------------------|----------------|-------------|------------------|
| $1\times10^{-2}$ | 650 | 0.5 | 14154282 | 1.231 | -1.117 | 1 | 2.48 |
| $1\times10^{-3}$ | 650 | 0.538 | 14154282 | 1.325 | -1.011 | 0.9 | 1.88 |
| $1\times10^{-4}$ | 650 | 0.691 | 14154282 | 1.701 | -1.296 | 0.8 | 1.67 |

The nonlinear refractive index of Rhodamine B for difference concentration was calculated (explain in table 4) from the transmission bend in the graph and by using eq. (14) for the close aperture Z-Scan technique, the results for rhodamine B dye showed exhibited self-defocusing (17,a). While the nonlinear absorption coefficient was calculated from eq.(18) from the transmission curve by using the open aperture Z-Scan and the results were explain in table(4). The result of the non-linear coefficient of absorption exhibited two photon absorption for the all concentration of Rhodamine B dye as shown in figure (17,b).

Figure (17) Z-scan technique experimental data for a. Close aperture and b. Open aperture

5. Conclusion

The experimental data of nonlinear optical properties of rhodamine B at different concentration explain that the sample has exhibited self-defocusing phenomena and negative non-linear refractive index while the nonlinear absorption coefficient exhibited a two-photon absorption this showed that the sample can be useful to use in nonlinear optical devices. Thereafter, the information we have found for optical properties is important showed that we could use this laser dye in optical limiter applications.
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