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Non-Linear and Linear Optical Properties of an Organic Laser Dye Mixture

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ABSTRACT

The present research includes studying the optical linear and nonlinear characteristics of two organic laser dyes: Nile Blue (NB), Malachite Green (MG) and their mixture at room temperature, a mixture proportions involving organic laser dye concentrations (2×10^{-5}, 4×10^{-5}, 6×10^{-5}, 8×10^{-5}) M in chloroform solvent was analyzed. Every produced sample has had its linear optical characteristics examined. Next, the UV-VIS spectra of various dye concentrations dissolved in absolute chloroform solvent have been examined. Finally, the absorption spectra of every sample have been examined. The results demonstrated that at the same wavelength, absorbance rose along with concentrations. The Z-scan technology was utilized to compute the nonlinear absorption coefficient (β) and nonlinear refractive index (n_2) for the produced samples in both open and closed aperture settings. The experiments were carried out using a solid-state laser with a diode pump that runs at 457 nm in wavelength and power of (84 mW). The results showed increasing of the nonlinear refractive index while declining nonlinear absorption coefficient for all organic laser dye samples at higher concentrations. Dyes mixture have large optical characteristics, both linear and nonlinear, in comparison to pure dyes.

1. INTRODUCTION

A dye laser is a laser in which the lasing medium is an organic dye, such as liquid solutions, gases, or solid state. Organic dyes are unsaturated hydrocarbon compounds with a broad absorption band in the visible range of the electromagnetic spectrum. Only organic compounds with an extended system of conjugated bonds alternating single and double bonds have this property [1, 2]. The transition from the electronic states for this process is usually very large, giving rise to an absorption bond ground state S_0 to the first excited singlet state, which is caused by long-wavelength of dyes absorption bond. The spontaneous emission in dye lasers is due to the reverse process S_1 to S_0 [3]. The physical phenomenon of energy transfer was first described over 90 years ago. Energy transfer is used to examine molecular level interaction because of its sensitivity to distance, as shown in Figure 1 [4].

The energy transfer rate of fluorescence emission has various applications in photochemistry, physics and biology. Another important use of energy transfer is in dye lasers, where it is used to reduce photo-quenching effects and thus increase laser efficiency [5]. Many studies are being performed in order to increase dye laser performance and broaden their operating spectral range. If two or three wavelengths are obtained simultaneously in a broadened region with only one dye system, use may be expanded.

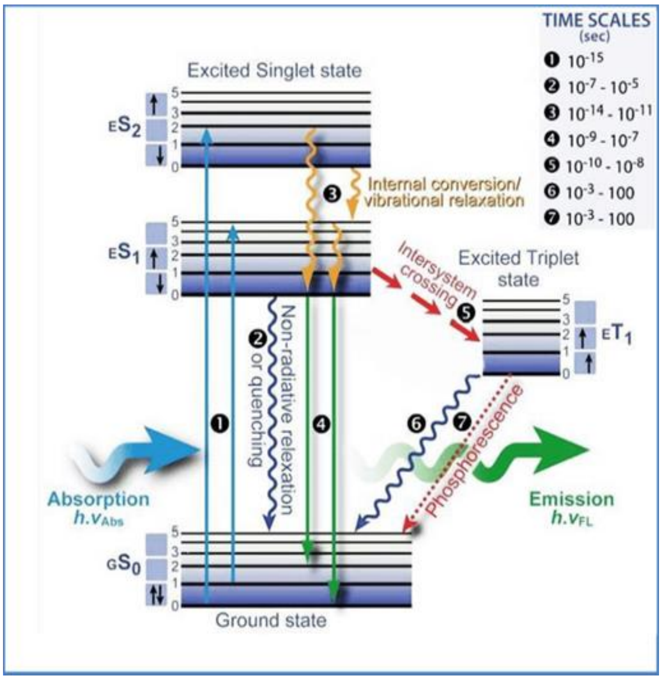

Figure 1. The Jablonski energy diagram shows the different energy states of the molecule and the possible transitions between them [4]

Dye mixtures are one of the easiest and most convenient ways to create such a laser. Earlier researchers detailed two-wavelength laser emissions from a two-dye mixture. One kind of dye mixture laser is an energy transfer dye laser (ETDL) [6]. The energy transfer mechanism is employed to provide superior outcomes, such as increased lasing efficiency and a greater spectrum of oscillations [7]. The aim of this research is studying the effect of mixing organic laser dyes to obtain a new dye with a wide range of absorption wavelengths in order to be used as effective media in dye lasers for multiple wavelengths and improving the linear and nonlinear optical properties of an organic laser dyes mixture, Nile Blue (NB) and Malachite Green (MG), which were dissolved in chloroform solvent at varying concentrations. Due to its broad applicability, Malachite Green was selected as the active medium in dye lasers; it is particularly prevalent in a variety of applications. Nile Blue, an oxazine dye, has also been the subject of our research.

2. LITERATURE REVIEW

Numerous researchers have produced a substantial quantity of data regarding laser dyes. Al-Aaraji et al. [8] examined the spectrum and linear optical characteristics of a blend of organic laser dyes Rhodamine B (RB) and Fluorescein Sodium (Na Fl) at varying concentrations. (10^{-3}, 10^{-4}) M in room temperature ethanol solvent. Comparing the combination of laser dyes to individual laser dyes, the results showed that the quantum efficiency decreased and the radiative and fluorescence life time rose with an increase in concentration. This indicates that the mixture of laser dyes is an effective optical material, and it can be used in cavity lasers as a resonator. Ali and Naser [9] investigated the linear optical characteristics of a combination of two organic laser dyes, phenol violet (10B) and rhodamine B (RB), dissolved in ethanol solvent at several concentrations in 2020. (2×10^{-6}, 4×10^{-6}, 6×10^{-6}, 8×10^{-6}) M at ambient temperature. All samples' absorption spectra were captured using UV-VIS. spectrophotometer. When compared to samples of separate laser dyes, the results showed that the mixed laser dye sample had far larger linear optical characteristics, and in dye lasers, it can be utilised as a laser active medium. Al-Hussainey et al. [10] investigated the non-linear optical characteristics of azure in 2022. An all-natural laser dye, dissolved with concentration in an ethanol solvent (10^{-4} M). The nonlinear optical characteristics were investigated in two cases —with a closed aperture and an open aperture—using the very sensitive Z-scan technique. in order to obtain two significant optical phenomena: nonlinear absorption coefficient and nonlinear refractive index. The outcome shows that the dye sample has two photon absorption (2PA) and a nonlinear refractive index that is negative. The results indicate that (Azure A) is a material with potential applications in nonlinear and photonic optical systems. According to the findings, every sample has potential for use in optoelectronic and photonic applications. The non-linear optical properties of organic laser dyes (Acriflavine, Orcein, and Azur-B) dissolved in ethanol solvent with concentration were investigated in 2023 by Khalil et al. [11] (10^{-5} M) were investigated in this study. The Z-scan method was utilised to ascertain the nonlinear optical characteristics in two distinct cases (close aperture and open aperture). This method is highly sensitive to determine the nonlinear refractive index and nonlinear absorption coefficient—the two fundamental optical phenomena. These dyes show promise as a material for photonic and nonlinear optical systems based on the results. Regarding the linear and nonlinear characteristics of a combination of malachite green and Nile blue dyes, no registered studies were discovered that addressed the same research topic. The nonlinear characteristics of Nile Blue, Malachite Green, and their mixing were made clear using Z-scan technology.

3. THEORY

The exploration of materials exhibiting rapid nonlinearities is a subject of considerable interest. This interest pertains to both nonlinear absorption and nonlinear refraction and is primarily motivated by looking for resources suitable for sensor protection and applications use just optical switching. The absorption of the material at high intensity is given by Eq. (1) [6]:

$$\alpha = \alpha_o + \beta I \tag{1}$$

where, (I) is the incident intensity, α_o is the intensity-dependent nonlinear absorption coefficient, and is the linear absorption coefficient. The refractive index at high intensity is determined by Eq. (2) [7]:

$$n = n_o + n_2 I \tag{2}$$

where, (n_2) is the nonlinear refraction coefficient and no is the linear refractive index.

The Z-scan technique is capable of determining the nonlinear absorption coefficient with an open aperture and when a closed-aperture shape is used, the nonlinear refractive index is employed to investigate nonlinear optical properties. Using the following Eq. (3) [8], the normalized transmittance's peak-to-valley difference is used to calculate the nonlinear refractive index:

$$n_2 = \frac{\Delta\Phi_o}{I_o L_{eff} k} \tag{3}$$

where, ($\Delta\Phi_o$) is the phase shift that is nonlinear [9]:

$$\Delta T_{p-v} = 0.406|\Delta\Phi_o| \tag{4}$$

The distinction between the normalized transmittances at peaks and valleys is denoted by ΔTp-v. Here, k = 2π/λ, where (λ) represents the spectrum of the beam, Io indicates the intensity level at the focal point, and Leff signifies the effective length of the sample. Al-Hussainey et al. [10] provide the following formula:

$$L_{eff} = \frac{(1 - \exp^{-\alpha_o L})}{\alpha_o} \tag{5}$$

where, (α_o) is the linear absorption coefficient, which is expressed as Eq. (6) [11]:

$$\alpha_o = \frac{\ln\left(\frac{1}{T}\right)}{t} \tag{6}$$

where, (t) represents the sample thickness as well as T denotes the transmission capacity. The index that is linear of refraction (n_o) as determined by Eq. (7) [12]:

$$n_o = \frac{1}{T} + \left[\left(\frac{1}{T^2} - 1\right)\right]^{1/2} \quad (7)$$

The nonlinear absorption coefficients (β) is a straightforward process using the subsequent Eq. (8) [13]:

$$\beta = \frac{2\sqrt{2}T(z)}{I_o L_{eff}} \quad (8)$$

T(z) is the lowest normalized transmittance value at the focus point, with z = 0.

4. MATERIALS AND CHEMICALS

This research was used in each of the following materials.

4.1 Nile blue

The Nile blue is a highly fluorescent oxazine dye [14, 15]. The NB was firstly synthesized by Möhlau and Uhlmann in 1896, molecular formula ($2C_{20}H_2ON_3O$). SO_4 and (MW = 732.85g/mol) were obtained from (Germany-SigmaAldrich), with a purity of 99.999%. Due to its environmental sensitivity, the cationic oxazine dye Nile blue has also been proven to be a solvatochromic dye. An electron acceptor (diethylamino group) and an electron donor make up the structure of this dye. Figure 2(a) shows the molecular structure of Nile Blue [16].

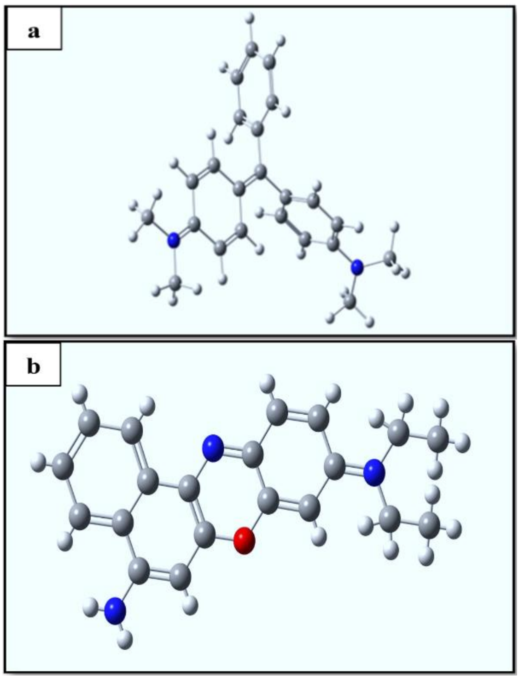

Figure 2. (a) Molecular Structure of Nile Blue dye; (b) Molecular Structure of Malachite Green dye.

4.2 Malachite green

Malachite green (chloride) is the dye employed in this research since it has several uses and serves as an active medium in dye lasers. As seen in Figure 2(b) [17], malachite green, also known as aniline green, Benz aldehyde green, or China green, has the chemical formula $C_{23}H_{25}ClN_2$ with a molar mass of 364.911g/mol.

4.3 Solvent

Chloroform was used as the solvent. It is relatively non-reactive, miscible with most organic liquids, chemical formula ($CHCl_3$), high purity (99.99%) of the highest purity available. It was used as a solvent for the laser dyes used in our current research.

4.4 Solution preparation

The solutions of concentrations (2×10^{-5}, 4×10^{-5}, 6×10^{-5}, 8×10^{-5}) M from each of two dyes Nile Blue and Malachite Green in Chloroform solvent were ready. An electronic balance with a four-digit sensitivity (BL210S), made in Germany, was used to weigh the powder. The following Eq. (9) was used to create various concentrations [18]:

$$W = \frac{M_W \times V \times C}{1000} \quad (9)$$

The mass (g), molecular mass (g/mol), solvent volume (mL), and concentration (C) of the dissolved material are denoted as W, Mw, V, and C, respectively [18]. The solutions that were prepared were diluted using the subsequent Eq. (10):

$$C_1V_1 = C_2V_2 \quad (10)$$

where, V_1 is the volume before dilution, V_2 is the amount after dilution, C_1 is the primary concentration, and C_2 is the new concentration.

4.5 UV-Visible spectroscopy

The linear optical properties including transmittance, absorption coefficient, and refractive index for all prepared samples have been determined by utilizing a UV-Visible Shimadzu 1800 Spectroscopy. This Spectroscopy is equipped with two light sources: a deuterium lamp and a tungsten lamp that operate in the wavelength ranges (190-390) nm and (390-1100) nm, respectively. All spectra were measured at room temperature in a quartz cell with (1 cm) optical path. as shown in Figure 3.

Figure 3. UV – Visible spectroscopy

4.6 Z-scan measurement

The Z-scan strategy is a straightforward and fundamental method for characterizing nonlinear absorption and refraction. It works using single beam technology. It refers to the process of passing a sample through a Gaussian beam focused in the central region. Wavefront disturbance caused by self-focusing or non-self-focusing will lead to nonlinearity in the Kerr scale. The power of the beam crossing a small aperture in the far field varies depending on the position of the sample. By estimating the output power with respect to the sample position, implemented using either methods with closed or open apertures, as shown in Figure 4 [19, 20].

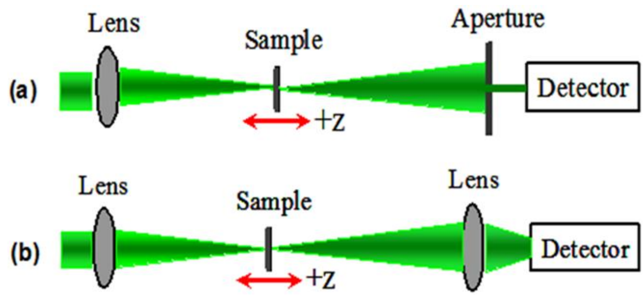

Figure 4. The setup of closed aperture Z-scan (a) closed aperture Z-scan and (b) open aperture zscan [21]

5. RESULT

5.1 Linear-optical properties

In this paper use two well-known laser dye groups: (NB) as an acceptor sample and (MG) as a donor sample. Different amounts of these two laser dyes were dissolved in chloroform solvent. (2×10^{-5}, 4×10^{-5}, 6×10^{-5}, and 8×10^{-5}) M. Absorbance spectra for (NB) dye and (MG) dye were measured and plotted as well as their mixture. The absorbance spectra of Malachite green dye at different concentrations as solutions were recorded in the region (250-750) nm as shown in (Figure 5).

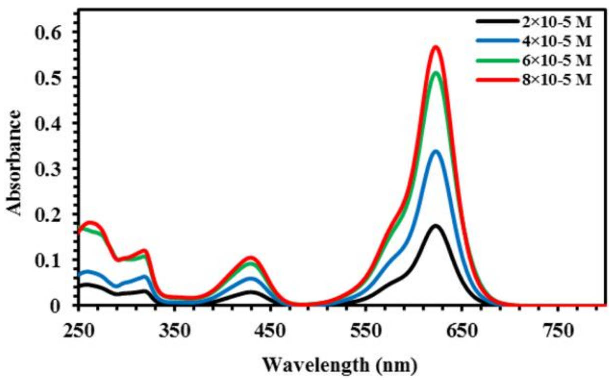

Figure 5. Malachite Green dye absorption spectra at various concentrations

This figure demonstrates that the four spectra include two bands: the Q-band, or visible area absorbance, which is located in the range of (420-620) increases strongly with concentrations increases according Beer-Lambert law, it agrees with the study of Mohammad et al. [6]. It was observed that the intensity of absorption is reduced by reducing the concentration and increase strongly with concentrations increases according Beer-Lambert law, it agrees with the study of Kumaresan and Ahamed [22].

We have also explored the potential use of Nile Blue dye in optical limiter devices. Nile Blue belongs to the family of oxazine dyes, which are well recognized for their remarkable resistance to thermal and photodegradation. Figure 6 illustrates the significant absorption band exhibited by Nile Blue at shorter wavelengths and in the red region above 600 nm, which is similar to the absorption characteristics of other dyes under study. Excitation occurs in the 400-550 nm range, where the original material is transparent. This transient absorption appears when a solution of Nile Blue in ethanol is excited at 400 nm with a pulse frequency of pulses per second. According to Beer-Lambert's law, absorbance increases with increasing concentration, leading to a decrease in transmittance, as it is inversely related to absorbance. The figure also shows that the width of the absorption spectrum narrows as the concentration increases.

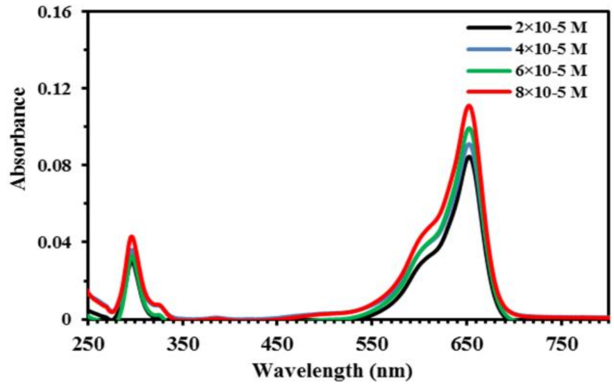

Figure 6. Absorption spectra for Nile Blue dye at different concentrations

The absorbance spectra of mixiture (NB + MG) organic laser dyes as solution at different concentrations as shown in Figure 7. The increase in the relative intensity of the mixture by increasing the concentration of the donor dye is due to the increased Probability for energy transfer and formation of the complex compound. In addition, it was found that the amount of spectral amplitude of the mixture increases with increasing concentrations of donor dye, which indicates an increase in the energy transfer efficiency, and it was found that the largest spectral range of the mixture at concentration (8×10^{-5}) M of the donor dye, i.e. an equal percentage of donor and acceptor molecules [19].

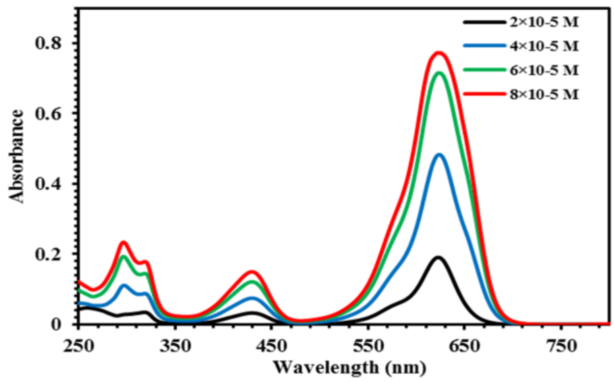

Figure 7. The absorbance spectra in chloroform solvent of two mixed dyes (NB and MG) at varying doses

5.2 Nonlinear optical properties

The Z-scan method was utilised to determine the nonlinear characteristics of Nile Blue dye, Malachite Green dye, and a mixture of (NB and MG) in chloroform solvent at different concentrations (2×10^{-5}, 4×10^{-5}, 6×10^{-5}, and 8×10^{-5}) M. The nonlinear absorption coefficient (β) is determined by open-aperture Z-scan, and closed-aperture Z-scan is used to calculate the coefficient of nonlinear refracted index (n_2). The dimensions were conducted at a power of (84 mW) and a wavelength of (457 nm).

Figures 8-10 illustrate closed-aperture Z-scans of the natural colour Nile Blue, Malachite Green dye, and a two-dye mixture at varying concentrations. There is an extension of the nonlinear effect zone from (-2) mm to (2) mm. The transmittance curve from the Z-scan data for closed apertures shows a peak and a valley, indicating a negative refraction nonlinearity ($n_2 < 0$), which causes self-defocusing lensing in the samples listed in Table 1, it agrees with the studies of Sinha and Dasgupta [23] and Ghanadan et al. [24]. To explain the Z-scan behaviour seen in the next Figures, note that the transmitted beam strength decreases and the transmittance essentially stays constant as the sample travels away from the focus.

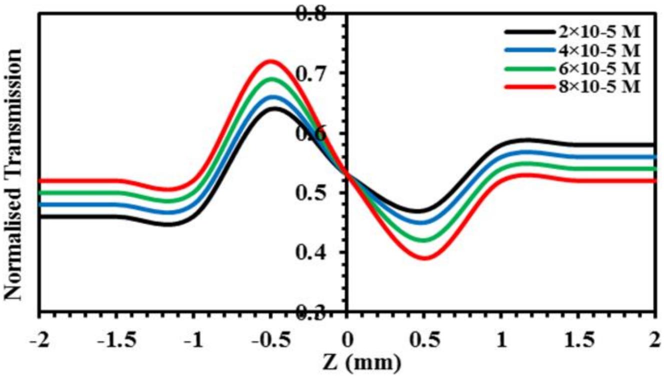

Figure 8. Z-scan with closed aperture used to measure the quantities of the dye Nile Blue sulphate

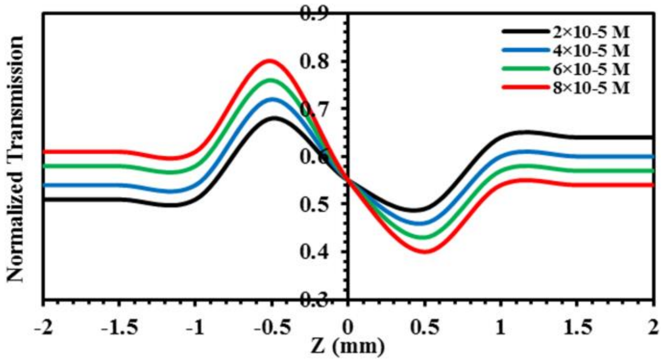

Figure 9. Z-scan with closed aperture used to measure the quantities of malachite green dye

In order to examine the nonlinear absorption coefficient, Open-aperture Z-scan of Nile Blue (NB), Malachite Green (MG) and mixing of them as solutions are shown in Figures 11-13. It has been observed that two photons absorb. The transmittance behaviour exhibits linearity at different distances from the far field of the sample point (-Z). In the adjacent field, Z = 0 mm at the focal point, the transmittance curve starts to decline and eventually approaches the minimal value (T min).

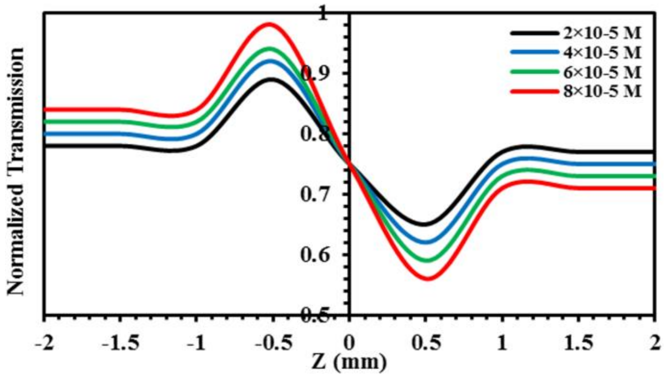

Figure 10. Z-scan with closed aperture of two mixture dyes (NB + MG) in chloroform solvent at varying concentrations

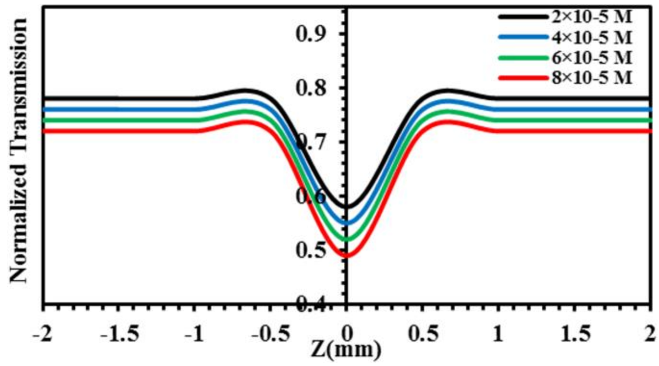

Figure 11. Z-scan of Nile Blue dye at various concentrations using an open aperture

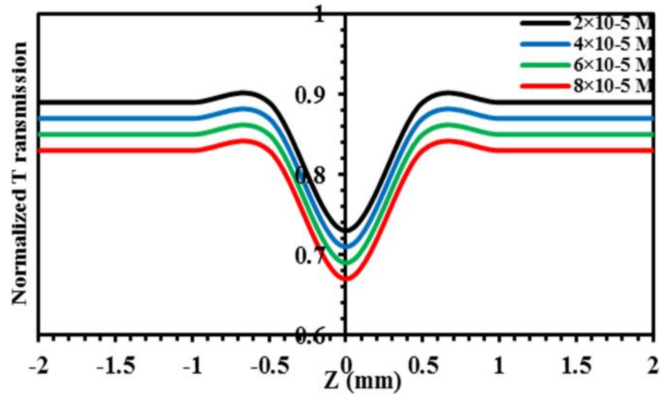

Figure 12. Open-aperture Z-scan of Malachite Green dye at different concentrations

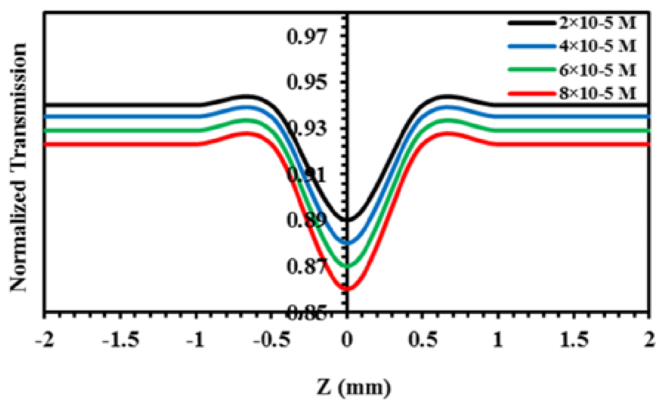

Figure 13. Z-scan data with an open aperture for a combination of dyes (NB and MG) at varying concentrations

Table 1. The optical characteristics, both linear and nonlinear, at various concentrations of Nile Blue, Malachite Green and Mixture of (NB + MG) at (λ = 457 nm)

Organic Laser Dyes	C Mol/L	T%	$\alpha_{\circ}$ cm^{-1}	$n_{\circ}$	ΔT_{p-v}	n_2 cm^2/mW	T(z)	β cm/mW
Nile Blue Dye	2×10^{-5}	1.0082	0.0027	0.9815	0.17	3.55×10^{-10}	0.58	1.91×10^{-3}
	4×10^{-5}	1.0057	0.0057	0.9871	0.21	4.39×10^{-10}	0.55	1.81×10^{-3}
	6×10^{-5}	0.9992	0.0075	1.0016	0.27	5.65×10^{-10}	0.52	1.71×10^{-3}
	8×10^{-5}	0.9972	0.0082	1.0061	0.33	6.91×10^{-10}	0.49	1.61×10^{-3}
Malachite Green Dye	2×10^{-5}	0.9865	0.0135	1.0306	0.19	3.98×10^{-10}	0.73	2.41×10^{-3}
	4×10^{-5}	0.9691	0.0313	1.0707	0.26	5.45×10^{-10}	0.71	2.34×10^{-3}
	6×10^{-5}	0.9483	0.0531	1.1196	0.33	6.92×10^{-10}	0.69	2.28×10^{-3}
	8×10^{-5}	0.9419	0.0598	1.1348	0.40	8.40×10^{-10}	0.67	2.21×10^{-3}
Mixture of (NB + MG)	2×10^{-5}	0.9846	0.015	1.035	0.24	5.03×10^{-10}	0.89	2.94×10^{-3}
	4×10^{-5}	0.9564	0.045	1.1005	0.30	6.29×10^{-10}	0.88	2.91×10^{-3}
	6×10^{-5}	0.9271	0.076	1.1700	0.35	7.35×10^{-10}	0.87	2.88×10^{-3}
	8×10^{-5}	0.9110	0.093	1.209	0.43	9.04×10^{-10}	0.86	2.85×10^{-3}

The transmittance starts to rise in the direction of the linear behaviour at the sample position's far field (+ Z). In this instance, two photon absorption occurs as the sample passes through the beam waist, which results in a shift in intensity. This behavior agrees with the study's point of Saleh et al. [25].

6. CONCLUSIONS

In this study, nonlinear optical properties of a mixture of Nile Blue, Malachite Green and Mixture of (NB + MG) Organic Laser Dyes were investigated as solutions dissolved in chloroform solvent. The nonlinear optical properties were ascertained utilizing the Z-scan technique, which is a highly sensitive method. A nonlinear absorption coefficient for two photons is observed across all samples. All of the samples demonstrate self-defocusing in their nonlinear refractive indices. As a result, energy transfer during more effective mixing from the donor dye to the acceptor dye, all mixture laser dye samples have significantly greater linear optical properties than individual laser dye samples. As an active laser medium, the mixture solution of these two dyes is more effective optically than the individual laser dyes. In photonic and nonlinear optical devices, the outcomes demonstrate that each sample comprises a material with great potential.

Using organic dyes, which include (Nile Blue) dye, (Malachite Green) dye, and (the mixture) as a solar concentrator to improve the performance of solar cells by benefiting from a large portion of the solar spectrum by the cell and converting it into electrical energy. Studying nonlinear optical properties of dyes using lasers with different wave lengths and different powers.

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