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# Spectral and Linear Optical Properties for New Mixture of Nile Blue and Malachite Green **Organic Laser Dyes**



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ABSTRACT

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### Keywords:

organic laser dyes, linear optical characteristics, mixture dyes, absorption spectrum

The aim of this study was to determine the spectral and linear optical properties for a mixture of Nile Blue (NB) and Malachite Green (MG) organic laser in chloroform solvent at different concentrations  $(2 \times 10^{-5}, 4 \times 10^{-5}, 6 \times 10^{-5})$  M. The measurements were conducted at ambient temperature. A study has been conducted on the energy transfer mechanism using Malachite Green (MG) as the donor and Nile Blue (NB) as the acceptor matrix. Examining fluorescence and UV-VIS absorption spectroscopy in the 500-700 nm wavelength region. The concentration and the absorption intensity are closely correlated. As the concentration went up, the quantum efficiency went down and fluorescence lifetimes went up. The findings indicate that using a blend of laser dyes is a more advantageous choice for optical materials in comparison to using single laser dyes.

# **1. INTRODUCTION**

Hydrocarbons are the classification for organic hues. Organic dyes are composed of complex, large molecules with an intricate structure. As you can see and infrared light waves hit them, they absorb and glow in a lot of different ways. In addition, their chemical weight is quite high. The carbon atoms in the linked chains that make up the chromophore system are joined by directionally shifting single and double bonds [1]. Anything that absorbs light in the visible and ultraviolet portions of the electromagnetic spectrum is called a chromophore. When the dye transitions in the visible range from its ground state  $(S_0)$  to its excited state  $(S_1)$ , light is absorbed by it. This is what gives the dye its color [2]. Chromophores are a category of compounds that provide color to a molecule. The dye molecule acquires a distinct color by selectively absorbing wavelengths within the range of 400 nm to 800 nm [3].

Laser dyes serve as the active medium in dye lasers, both pulsed and continuous wave (CW). Laser dyes can be either solid or liquid. They are also used in rapid shutters for inactive mode locking and Q-switching. Hence, it is necessary to possess a diverse array of dyes at one's disposal in order to encompass the whole of the spectrum [4].

Tememee and Habeeb [5] studied the spectral properties of mixed Coumarin and Rhodamine dyes at different solvent effect of their properties. The results showed that, increase the intensity of absorption for both dye with increase of the dye concentration, also, shifted the wavelength to longer (red The maximum efficiency was 94% shift). for (Rhodamine+Coumarin) mixtures in ethanol solvent.

Kadhum and Abbas [6] studied the effect of fluorescence

energy transfer between laser organic dyes. It included preparing a mixture model consisting of Aniline blue dye as (donor) with a Malachite Green dye as (accepter). Furthermore, a significant overlap between the absorption and fluorescence spectra of the two dyes is obtained, which validates the accepter + donor hypothesis. First, the two dyes' absorption and emission (fluorescence) spectra were examined at four distinct concentrations that were dissolved in ethanol solvent. The results show that the decrease in concentration leads to a decrease in the intensity of absorption and emission (fluorescence) for all the prepared samples.

According to Abdulrazzaq et al. [7], the development of dye lasers is specifically dependent on the considerable and effective phenomenon of excitation energy transfer between dye molecules, which creates the ideal operating conditions for lasing at desired wavelengths. At ambient temperature, at various concentrations  $(2 \times 10^{-5}, 4 \times 10^{-5}, 6 \times 10^{-5}, and 8 \times 10^{-5})$ M in ethanol solvent, the linear optical characteristics of a combination of Rhodamine B (RB) and Malachite Green (MG) organic laser dyes were assessed both before and after mixing. With increasing concentration, the mixture showed increases in its refractive index, broad absorption spectral bandwidth, and linear absorbance index. Depending on the concentrations of the donor sample, the highest absorption bandwidths in the combination, which ranged from 470 to 650 nm, were achieved at concentrations of  $8 \times 10^{-5}$ ) M for each dye and a mixing ratio of 1:1. This series offers a large-scale laser pumping source.

Abdulridha and Naser [8] investigated the optical characteristics of a PMMA polymer doped with Malachite Green organic laser dye. Cu nanoparticles were examined at concentrations of (2×10<sup>-5</sup>, 4×10<sup>-5</sup>, 6×10<sup>-5</sup>, and 8×10<sup>-5</sup>) M in chloroform solvent. A UV-VIS spectrophotometer was used to record each sample's transmission and absorption spectra. In contrast to dye-doped PMMA polymer samples, samples of pure organic dye exhibit lower linear optical properties. This is demonstrated using Cu nanoparticles doped with organic color. This finding demonstrated how highly suitable these organic dyes doped with nanoparticles are for applications involving optical and photonic devices.

Fahdel Odah et al. [9] studied how to modify the spectroscopic properties of Nile Blue laser dye using MgO and NiO nano oxides. A suitable amount of ethanol was used to dissolve the powdered Nile Blue laser dye in order to produce a solution with different concentrations, such as (10<sup>-1</sup>, 10<sup>-2</sup>, 10<sup>-</sup> <sup>3</sup>, 10<sup>-4</sup>, and 10<sup>-5</sup>) mol/l. The structural characteristics of two metal nano-oxides, NiO and MgO, were investigated using XRD and FE-SEM methods after they were chemically produced as suspensions. Four distinct quantities of NiO and MgO NP suspension were mixed with the laser dye solution (0.5, 1, 1.5, and 2 mol/l). The resulting pure dye laser solution's absorption and fluorescence spectra for the combination were then examined. Quantitative yield, Stockes shift, FWHM, fluorescence, maximum wavelength maximum of fluorescence, fluorescence life duration, and Stockes shift were among the other fluorescence characteristics that were determined.

The current work aims to investigate the linear and spectral characteristics of a combination of two organic laser dyes. The aim of combining two laser dyes is to achieve new qualities without having to produce dyes with those features on their own. It also helps to identify potential industrial uses for the new properties that result from the combination of these two dyes. According to the results, these dyes might be used as building blocks for linear and photonic optical systems. As far as the spectrum and linear properties of a blend of Nile Blue and Malachite Green pigments are concerned, no registered papers that addressed the same study issue have been found.

# 2. MATERIALS AND CHEMICALS

A very luminous oxazine dye is Nile Blue [10, 11]. Nile Blue is a well-performing organic dye that is extremely fluorescent and photostable. It is a member of the a Benzophenoxazine family. The self-association feature of this dye in stock solution, as well as its many uses in the medical area, particularly in laser dye, has piqued my curiosity. The molecular formula is  $2C_{20}H_2ON_3O$ . The compound SO<sub>4</sub>, weighing 732.85 g/mol, which was purchased from SigmaAldrich in Germany. It has a purity of 99.999%. Figure 1(a) displays the molecular structures of NB organic laser dye.

Malachite Green, or chloride, is the color utilized in this composition. It was chosen for the dye laser due to its many applications and efficacy as a medium. As can be observed in Figure 1(b), aniline green, benz aldehyde green, China green, and Malachite Green all share the same formula,  $C_{23}H_{25}ClN_2$ , and a molar mass of 364.911 g/mol [7].

#### 2.1 Solutions preparation

Highly concentrated solutions (10<sup>-3</sup> M) of each organic laser dye were prepared using chloroform as the solvent. A German electric balance model (BL 210 S) with a resolution of four decimal places was used to measure the powder. Equation was used to make different concentrations [12, 13]:

$$W = \frac{M_W \times V \times C}{1000} \tag{1}$$

where, W is molecular weight, measured in grams per mole, V, in milliliters, and its amount, C, are all given. This solution [14] was used to water down the results that were already given:

$$C_1 V_1 = C_2 V_2 \tag{2}$$

These are the locations: C<sub>1</sub> and C<sub>2</sub> indicate the first and second concentrations, respectively. It is crucial to remember that V<sub>1</sub> represents the volume before to diluting and V<sub>2</sub> represents the volume following diluting. In this work, four concentrations (2×10<sup>-5</sup>, 4×10<sup>-5</sup>, 6×10<sup>-5</sup>, and 8×10<sup>-5</sup>) M of each of the dyes Malachite Green (MG) and Nile Blue (NB) were used to prepare a mixture of these two dyes 1 mL of Malachite Green (MG) and 1 mL of Nile Blue (NB) to prepare a mixture of them in different concentrations (2×10<sup>-5</sup>, 4×10<sup>-5</sup>, 6×10<sup>-5</sup>, and 8×10<sup>-5</sup>) M, as shown in Figure 2.



Figure 1. (a) Molecular structure of NB organic laser dye; (b) Molecular structure of MG organic laser dye



Figure 2. (a) NB organic laser dye, (b) MG organic laser dye and (c) Mixture of (NB +MG) organic laser dyes at diffirent concentration

### 2.2 Devices used

#### 2.2.1 UV-visible spectroscopy

The linear optical characteristics, such as transmittance, absorption coefficient, and refractive index, of each produced sample were determined using a UV-visible Shimadzu 1800 Spectroscopy. This spectroscopy uses a deuterium lamp and a tungsten lamp as its two light sources. which operate in the wavelength ranges of 390-1100 nm and 190-390 nm, respectively. Computer software is utilized to compute the optical constants using the output data on wavelength, transmittance, and absorbance. Figure 3 illustrates that the detector is a silicon photodiode.



Figure 3. UV-visible spectroscopy

#### 2.2.2 Fluorescence measurement

Fluorescence spectra was measured for all the prepared samples using spectrometer type (FluoroMate FS-2). Fluorescence emission was measured from the samples prepared at the section using spectro fluorophotometer type of (RF-5301pc Shimadzu) the sample were mounted cubic cell of quartz dimensions  $(1 \times 1 \times 5)$  cm at angle (90°) with incident beam. This optical geometry was chosen to eliminate the effect of scattered incident radiation. The instrument computerized and operates in the wavelength range (220-900) nm. The fluorimeter has dedicated computer which control instrumental operating (excitation and emission wavelength, scan, monochromator slit width, detector parameter) and the acquisition of spectral data, as shown in Figure 4.



Figure 4. Fluorescence spectroscopy

# **3. EXPERIMENTAL WORK**

The formula was used to obtain the linear absorption coefficient  $(\alpha_0)$  [13].

$$\alpha_o = \frac{\ln\left(\frac{1}{T}\right)}{t} \tag{3}$$

The sample's thickness is shown by t, its transmittance by T, and its refractive index by  $n_o$ . The formula [14] may be used to ascertain the existence of the film based on its transmittance spectrum.

$$\mathbf{n}_{o} = \frac{1}{T} + \left[ \left( \frac{1}{T^{2}} - 1 \right) \right]^{1/2} \tag{4}$$

Transmittance refers to the percentage of light that is able to pass through a solution. Therefore, when half of the light passes through, it may be said that the solution has a transmittance of 50% [14]:

$$T\% = \left(\frac{l}{I_{\circ}}\right) \times 100\% \tag{5}$$

where,  $I_{\circ}$  is the strength of the light beam coming in and I is the strength of the light that the liquid is giving off. You can use math to show how the factors transparency T and absorption A are related by the equation [13,15].

$$A = \log_{10}\left(\frac{I_{\circ}}{I}\right) \tag{6}$$

Using the Formula, one may calculate the fluorescence quantum yield  $\Phi_f$  and the fluorescence lifetime based on the findings of the fluorescence spectra [15].

$$\tau_f = \frac{a \times \tau_{fRB}}{a_{RB}} \tag{7}$$

The lifetime of the standard chemical, denoted as  $(\tau_{fRB})$ , The measurement is determined by calculating the integral of the fluorescence curve of the laser organic dye [15].

$$\phi_f = \frac{\int F(v') \, dv'}{\int \varepsilon(v') \, dv'} \tag{8}$$

The area under the curve is denoted by  $\int F(v') dv'$ , while the area under the absorption curve is denoted by  $\int \varepsilon(v') dv'$ .

### 4. RESULT AND DISCUSSION

Two well-known sets of laser dyes were used in the experiment: MG served as the donor and NB as the acceptor.  $2 \times 10^{-5}$ ,  $4 \times 10^{-5}$ ,  $6 \times 10^{-5}$ , and  $8 \times 10^{-5}$  M of two laser dyes were dissolved in a chloroform solvent. The combination solutions including NB, MG, and a mixture of NB and MG had their absorbance spectra and absorbance values recorded and displayed. Additionally, the quantum yield and fluorescence lifetime were ascertained. The Shimadzu 1800 Spectrophotometer and the FluoroMate FS-2 Fluorescence Spectrophotometer were used to record the UV-Vis absorption and fluorescence spectra of the dye laser solutions, respectively.

#### 4.1 The absorption spectra



Figure 5. UV-VIS absorption spectra for NB organic laser dye at different concentrations

The absorption spectra of the NB organic laser dye, when it is dissolved in chloroform at various concentrations  $(2 \times 10^{-5}, 4 \times 10^{-5}, 6 \times 10^{-5}, and 8 \times 10^{-5})$  M, were measured throughout the

wavelength range of 400-800 nm. These results are shown in Figure 5. The absorption spectra of the substance are reported to have a rather wide range between 462 and 700 nm. It has been noted that the strength of the absorption spectra increases as the concentration increases, in accordance with Beer-Lambert's equation.

Figure 6 shows the absorbance spectra of the MG organic laser dye at different concentrations, with solutions measured between 400 and 800 nm. As can be seen in this figure, the four spectra show two different bands: the visible area absorbance, or Q-band, which is within a specific range. According to Beer-Lambert law, (400-700) grows significantly as concentrations rise [7].



Figure 6. UV-VIS absorption spectra for MG organic laser dye at different concentrations



**Figure 7.** UV-VIS absorption spectra for Mixture of (NB+MG) organic laser dyes at different concentrations

Figure 7 displays the absorbance spectra of a mixture of (NB + MG) organic laser dyes in solution at different concentrations. The current findings demonstrate that the absorption peaks exhibited a shift towards longer wavelengths as the concentrations increased. This shift occurs as a result of the increased density of molecules at high concentrations. The figure illustrates that the four spectra exhibit two distinct bands in the ultraviolet (UV) area, specifically referred to as the B-band, with a wavelength range of around 400-470 nm. These bands arise from electronic transitions involving the movement of electrons from  $\pi$  orbitals to  $\pi^*$  orbitals. The Q-band, located in the visible region, spans a range of around 500-700 nm and is characterized by the longest wavelengths. This band corresponds to the transition from the n orbital to the  $\pi^*$  orbital. The absorbance also increased as a result of

increasing the concentration of the mixture, leading to a higher number of molecules per unit volume. This affects the energy state, which is consistent with reference [7]. In addition, the absorbance of a mixture of organic laser dyes, specifically NB and MG, is significantly greater than that of the individual dyes. This phenomenon may be explained by the occurrence of energy transfer between the molecules that donate and receive energy.

### 4.2 The Fluorescence Spectra

The NB dye diluted in chloroform at different concentrations (2×10-5, 4×10-5, 6×10-5, and 8×10-5) M is shown to have a fluorescence spectrum in Figure 8 at a wavelength of 600- 850 nm. When molecules are present, the energy they absorb to move back to their original positions creates the displaced fluorescence spectrum, also known as the Stokes Shift, which is visible at infrared wavelengths. This energy is called relaxation energy [15]. It is seen that when the concentration increases, there is a corresponding increase in the fluorescence intensity and a shift towards longer wavelengths (red shift). This is attributed to the interaction between the particles of the medium when the concentration varies [16]. The fluorescence lifetime  $(\tau_f)$  and quantum yield of fluorescence  $(\Phi_f)$  were calculated using the absorbance and fluorescence spectra data. It is observed that higher concentration levels result in an elevation in fluorescence lifetime  $(\tau_f)$  values and a reduction in fluorescence quantum yield ( $\Phi_f$ ) values, as depicted in Table 1.



Figure 8. Fluorescence spectra for NB organic laser dye at different concentrations

Figure 9 shows fluorescence intensity for MG organic laser dye at different concentrations. From this figure, it is noticed

that the fluorescence intensity increases with further increase in concentration, it agrees with references [17].



Figure 9. Fluorescence spectra for MG organic laser dye at different concentrations

Figure 10 displays the fluorescence intensity when (NB+MG) organic laser dyes are mixed at concentrations of  $(2 \times 10^{-5}, 4 \times 10^{-5}, 6 \times 10^{-5} \text{ and } 8 \times 10^{-5})$  M as solutions. At larger concentrations, fluorescence inspection of these spectra revealed a red shift in the emission peaks, indicating a shift towards longer wavelengths. The shift occurred as a result of the higher concentration of molecules per unit volume [15, 17].



Figure 10. Fluorescence spectra for Mixture for (NB+MG) organic laser dyes at different concentrations

Table 1. \	Values of	Fluorescence	and Optical	Properties for	r Pure and Mixtu	re dyes at	t different	concentrations
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Materials	Concentration (M)	λ <sub>max</sub> (Absorbance)	α. (cm <sup>-1</sup> )	n∘	$\lambda_{max}$ (Fluorescence)	$\tau_{f}$ (ns)	$\Phi_{\mathbf{f}}(\mathbf{\%})$
	2×10 <sup>-5</sup>	652	0.002	0.983	680	0.122	82
ND anamia la san dara	4×10 <sup>-5</sup>	652	0.005	0.988	680	0.126	80
NB organic laser dye	6×10 <sup>-5</sup>	652	0.007	1.001	680	0.133	78
	8×10 <sup>-5</sup>	652	0.008	1.005	680	0.136	76
	2×10-5	623	0.013	1.027	674	0.117	88
	4×10 <sup>-5</sup>	623	0.031	1.064	674	0.118	85
MG organic laser dye	6×10 <sup>-5</sup>	623	0.053	1.110	674	0.120	83
	8×10 <sup>-5</sup>	623	0.059	1.125	674	0.122	82
	2×10-5	623	0.015	1.092	673	0.128	99
Mixture of (NB+MG) organic laser dyes	4×10 <sup>-5</sup>	623	0.045	1.193	673	0.150	97
	6×10 <sup>-5</sup>	623	0.076	1.218	673	0.158	95
	8×10 <sup>-5</sup>	623	0.093	1.483	673	0.163	93

For the donor NB organic laser dye molecule to transmit energy to the acceptor molecule, there must be spectral overlap between their absorption and fluorescence spectra. The amount of overlap in the spectrum determines how efficiently energy is transmitted. The two spectra are seen to overlap rather well in Figure 11. This overlap demonstrates the extremely high transmission efficiency of energy between both wavelengths. If the acceptor's absorption spectra and the donor's fluorescence spectrum are not complementary, energy cannot be transferred from the donor to the acceptor molecule. References [18, 19] support the idea that energy transmission efficiency is influenced by the degree of spectral concordance.



Figure 11. Spectral overlap between NB's absorbance and MG's fluorescence spectra at  $(8 \times 10^{-5})$  M

#### 4.3 Calculation of quantum efficiency and life time

Table 1 displays the time-life values and quantitative efficiency of all prepared samples. The time-life and quantitative efficiency were determined using Eqs. (7) and (8), respectively. In order to do the measurement, the area under the fluorescence curve divided by the area under the absorbance curve was determined using the computer program GEUP 9. The results showed that the highest quantitative efficiency at concentration  $(2 \times 10^{-5})$  of mixture dyes (NB+MG) When the concentration of organic laser dyes increases, the quantitative efficiency decreases. The reason is because the quantitative efficiency is the ratio between the area under the curve of the fluorescence to the area under the curve for absorption. When the concentration increases, the absorbance increases according to Beer-Lambert's law, and in case of increasing the absorbance, the quantitative efficiency decreases. It has been demonstrated that for two distinct kinds of dye lasers (NBA+MG), as concentration grew, so did the absorption and fluorescence intensity at the maximum wavelength. This is because to the high intermolecular distance and low concentration that the dye molecules adopted. The observed fluorescence spectrum is impacted by this process in addition to red shifting, or wavelength displacement in the peak position, which is attributed to selfabsorption. This is because the absorption spectrum overlaps with the long wavelength region, increasing its intensity, and the short wavelength region decreases its intensity [20, 21].

# **5. CONCLUSIONS**

The study concludes that the absorption spectra of both dyes have shown a rise as the concentrations grow. Additionally, the fluorescence spectra exhibit a positive correlation with the concentrations, meaning that as the concentrations increase, the fluorescence spectra likewise increase. Because of the extended radiative and fluorescence lifetimes, the quantum efficiency dropped as the concentration increased. All of the combined laser dye samples exhibit significant spectrum and linear optical properties when compared to samples of pure laser dyes, according to the results. The mixed laser dye solution's fluorescence spectra at concentration ( $8 \times 10^{-5}$ ) M are greater than those at other concentrations, making it suitable for use as an active laser medium.

### **6. FUTURE WORKS**

• Analysing how a combination of several laser dyes' linear and nonlinear optical characteristics are affected by pulsed lasers with nanosecond and femtosecond durations.

• Designing effective laser media using laser dyes dispersed within nanoparticles at calculated concentrations.

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