

Optical Nonlinearity and Optical Limiting Performance of Disperse Blue 1/Dioxane via Z-Scan Technique

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Abstract

The absorption performance and evolution of the organic dye disperse blue 1 (DB1) in dioxane at different concentrations was investigated. The nonlinear absorption coefficient has been measured through the open aperture Z-scan technique, whose values increased exponentially from 0.997×10^{-3} to 11.7×10^{-3} cm/W as the dye concentration increased from 3.7×10^{-5} to 74.6×10^{-5} M. Also, DB1 optical power limiting characteristics appeared at high laser light intensities, whereby very dilute solution of the order of $\sim 10^{-4}$ M was sufficient to limit the transmitted power of the incident laser light by $\sim 90\%$.

Keywords: Nonlinear Absorption, Disperse Blue 1, Z-Scan, Optical Limiting

1. Introduction

For the last few decades, Organic dye materials with enhanced nonlinear optical (NLO) properties have been an active field of research for wide applications in all-optical switching, optical communications, optical power limiting, optoelectronic and photonic devices; owing to the growing needs in eye and optical sensor protection from intense laser exposure, as well as for protective applications in optical sensors.¹⁻⁹

Organic dyes have been recognized as important materials for optical applications owing to their large third-order nonlinearity, photochemical stability, fast response time, high damage thresholds, applicability over a wide range of the visible spectral region, and easy fabrication into optical devices.¹⁰⁻¹³ For optical power limiting applications, the transmittance of such optical materials is desired to be inherently nonlinear, *i.e.*, the materials should be highly transparent at low input energies while the transmittance has to drop with increasing the input energy. Fullerene materials displayed potential optical limiting properties since the early days of their discovery in 1985.^{14,15} Tutt *et al.* investigated the optical limiting of C₆₀ and C₇₀ solutions using 8-ns pulses of 532 nm laser light.¹⁵ They found that the strong absorption behavior in these materials with saturation thresholds was equal or lower than other materials reported to that date. Noble metal nanoparticles

incorporated in polymer matrices forms another promising field in the quest for efficient optical limiting materials. Porel *et al.*¹⁶ have embedded gold and silver nanoparticles into poly(vinyl alcohol) films, which showed a limiting threshold of ~ 0.7 J.cm⁻², at 800 nm Ti:Sapphire laser (~ 100 fs). Semiconductor nanoparticles form another interesting class of promising materials due to their unique quantum nature, which changes the material solid-state properties. Irimpan *et al.* studied the spectral and nonlinear optical properties of ZnO-CdS nanocomposites prepared via colloidal chemical synthesis, using nanosecond laser pulses at the excitonic resonance and off-resonance wavelengths.¹⁷ They observed that the nonlinear response is wavelength dependent, and switching from saturable absorption (SA) to reverse SA (RSA) for samples as the excitation wavelength changes from the excitonic resonance to off-resonance wavelengths. Nagaraja *et al.*¹⁸ studied the effects of annealing on the third-order optical nonlinearity and optical power limiting properties of sputtered MnO and ZnO thin films in CW laser regime investigated by Z-scan technique. They found that the incorporation of Mn into ZnO and annealing lead to prominent changes in the third order nonlinearity.

The present study focuses on the nonlinear optical absorption (NLA) and optical power limiting properties of Disperse Blue 1/dioxane solution at different

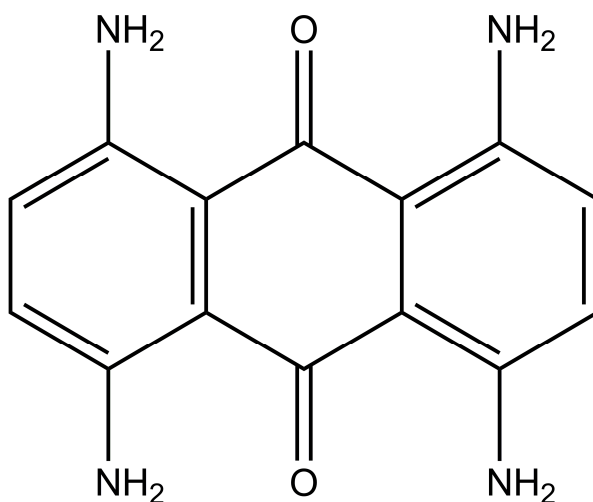


Fig. 1: Molecular structure of DB1 dye.

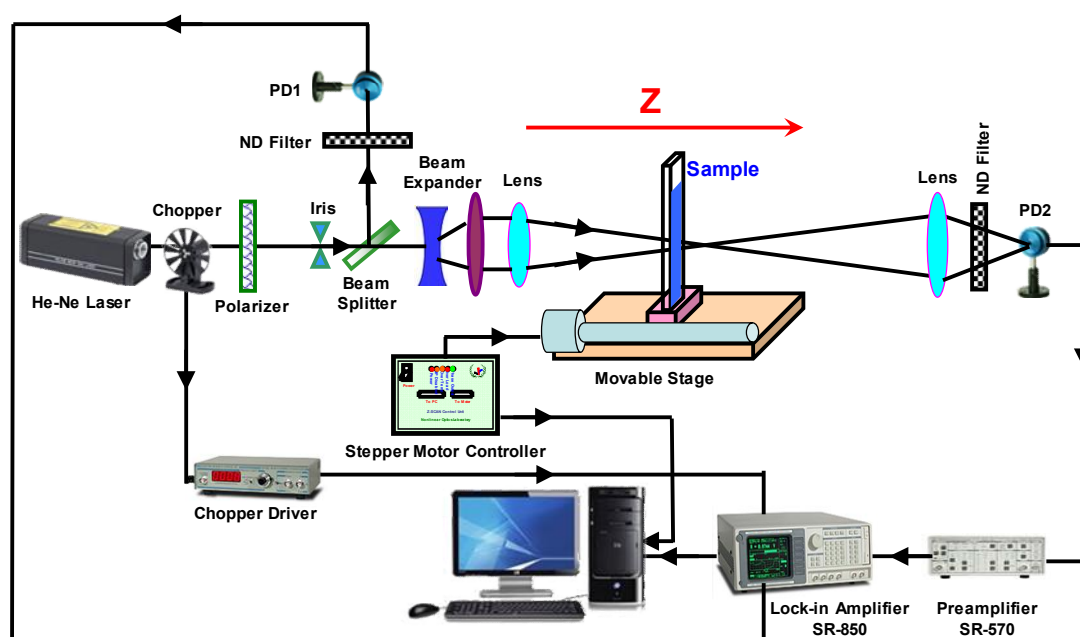


Fig. 2: Schematic drawing of the automated Z-scan system (open aperture configuration) for measuring the nonlinear optical absorption coefficient.

concentrations, under the influence of CW laser radiation at 632.8 nm. The chosen Disperse Blue 1 (DB1) dye has never been studied before - to the best of our knowledge - and is a promising candidate for optical power limiting and other optical devices due to its strong nonlinear response at visible region wavelengths, especially at the wavelength of 632.8 nm.

2. Experimental

2.1. Sample Preparation

Disperse blue 1, DB1, ($C_{14}H_{12}N_4O_2$, MW: 268.28 g/mol) was purchased from Acros Organics. Fig. 1 shows the molecular structure of DB1 dye. Solutions of DB1 in dioxane were prepared by solving the 0.1 gr of DB1 in 20 ml of 1,4-dioxane, and mechanically stirred for four hours in a closed flask at room temperature. A series of different concentrations (3.70×10^{-5} , 7.46×10^{-5} , 1.12×10^{-4} , 1.49×10^{-4} , 1.86×10^{-4} , 3.73×10^{-4} , and 7.46×10^{-4} M) were prepared from this solution using a precise digital

micropipette (Transferpette Electronic, from Brand). The test cell used in this work was 1 mm thick quartz cell.

2.2. Experimental Setup and Instrumentation

A CW high power linearly polarized TEM₀₀ Gaussian beam He-Ne laser (31-2140-000, from Coherent) was used in this work, with a wavelength of 632.8 nm. The optical setup used for Z-scan measurements is shown in Fig. 2. A slight portion of the laser beam was reflected by a microscope slide and directed towards the photodiode PD1, which was used as a reference beam for normalizing the collected data throughout the experiments. The laser beam was focused to about 31 μ m of waist radius with a convex lens ($f = 150$ mm), at which the Rayleigh length Z_R was 0.46 cm. Hence, the calculated power density was ~ 2.4 kW/cm² at the focus.

The sample was mounted on a home-made Z-scan mechanical assembly, which was controlled by the software via a stepper motor controller. The transmitted

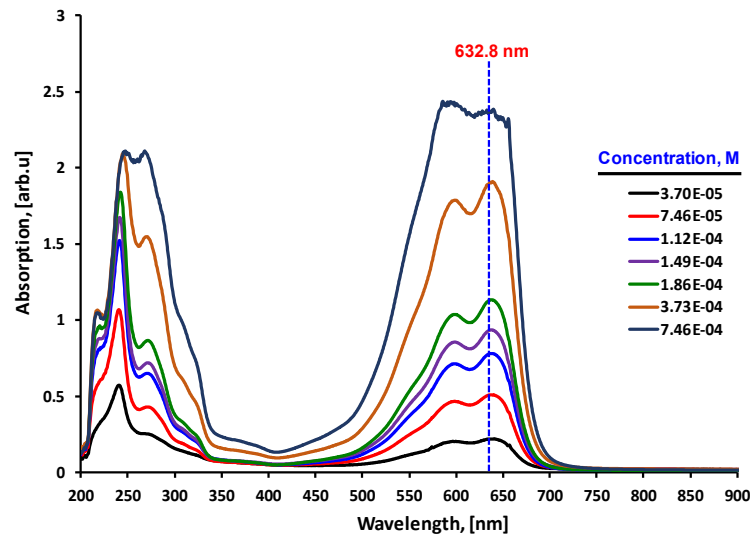


Fig. 3: UV-visible absorption spectra of DB1/dioxane solutions at different concentrations.

Table 1: Summary of the calculated parameters for DB1/dioxane solution at different concentrations.

Concentration $\times 10^{-5}$ [M]	α [cm ⁻¹]	$L_{eff} \times 10^{-2}$ [cm]	$\beta \times 10^{-3}$ [cm/W]	$\gamma \times 10^{-8}$ [cm ² /W]	$\sigma_{gr} \times 10^{-16}$ [cm ²]	$\sigma_{exc} \times 10^{-3}$ [cm ²]	OL Intensity Threshold [mW]
3.7	5.04	7.86	0.997	-5.69	2.26	0.1	20.4
7.46	7.6	7.01	1.6	-5.56	1.69	1.56	8.5
11.2	17.83	4.67	3.51	-4.86	2.64	3.51	7.7
14.9	21.3	4.14	4.57	-4.18	2.37	4.57	7
18.6	25.83	3.58	5.5	-3.70	2.31	5.5	0.76
37.3	43.53	2.27	9.22	-3.53	1.94	9.22	0.0496
74.6	53.46	1.86	11.73	-3.40	1.19	11.73	0.0017

beam was collected by a lens ($f = 75$ mm), and incident on the photodiode PD2. The intensity of the reference beam (through the photodiode PD1) was collected and recorded via the auxiliary input of the DSP lock-in amplifier (SR-850, from Stanford Research Systems), whereas the intensity of the sample beam (through photodiode PD2) was collected and recorded via a current preamplifier (SR-570, from Stanford Research Systems), which was connected to the DSP lock-in amplifier. A chopper (SR-540, from Stanford Research Systems) was connected to the lock-in amplifier for the signal detection. An IEEE 488.2 GPIB (from National Instruments) card was used to control and record the experimental data along with a special program written in Borland C++. The Lab-Master Ultima Coherent power meter was used to measure the incident power of the laser beam.

3. Results and Discussions

3.1. Absorption Coefficient Calculations

A UV-visible spectrophotometer (Photodiode Array Photospectrometer (PDA) Specord S100, from Analytik Jena) was used to record the UV-visible absorption spectra of DB1 liquid samples. Fig. 3 shows the absorption dye spectra of the prepared concentrations. The absorption increased as the dye concentration did, and a saturation effect started to appear in the highest concentration used. This figure showed as well the irradiation laser wavelength position relative to the absorption maxima.

Fig. 3 is about here

The linear absorption coefficients of the samples at 632.8 nm were calculated using the Beer-Lambert's law,¹⁹

$$\alpha = \frac{\ln\left(\frac{1}{T}\right)}{L} \quad (1)$$

Where T is the sample's transmission at 632.8 nm, and L is the sample thickness. The effective path length of the laser beam inside the sample was deduced from the results of the linear absorption coefficient values, using the formula,²⁰

$$L_{eff} = \frac{1 - e^{-\alpha L}}{\alpha} \quad (2)$$

Table 1 lists the calculated values of both linear absorption coefficients and the effective path lengths for all concentrations used.

3.2. Open Aperture Z-Scan Measurements

The nonlinear absorption is a result of either the direct multiphoton absorption or the saturation of single photon absorption.²¹ The open aperture Z-scan was expected to be symmetric with respect to the focus ($z = 0$) where it had the minimum transmittance (for two or multiphoton absorption) or maximum transmittance (for absorption saturation). A typical result of the Z-scan measurements is shown in Fig. 4, which corresponds to the normalized transmission of the DB1/dioxane sample as a function of

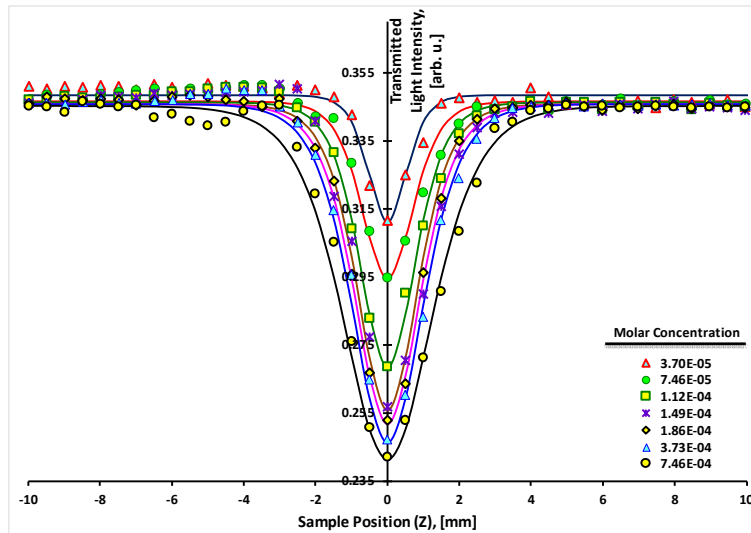


Fig. 4: Normalized transmittance curves of open aperture Z-scan for DB1/dioxane solution at different concentrations.

its distance (Z) from the lens focus. The solid lines display the theoretical fits.

The nonlinear absorption coefficient β was estimated from the open aperture Z-scan data, using the following formula,²⁰

$$T(z) = \sum_{m=0}^{\infty} \frac{[-q_0(z)]^m}{(m+1)^{(3/2)}} \quad (3)$$

For $q_0 < 1$; where $T(z)$ is the normalized transmittance, $q_0 = \frac{\beta I_0 L_{eff}}{Z_R^2}$, $Z_R = k \omega_0^2 / 2$ is the

beam's diffraction length, ω_0 is the beam waist radius at the focal point, $I_0 = \frac{2P_0}{\pi \omega_0^2}$ is the intensity of the laser

beam at focus, P_0 is the laser input power, and $k = 2\pi / \lambda$ is the wavevector.

These normalized transmission curves were characterized by the focal valleys. Moreover, the depths of these valleys increased with increasing the molar concentrations of the samples, and a minimum transmittance at $z = 0$ for all samples was evident, which was in agreement with the two-photon absorption (TPA) process.²¹ However, one must not forget the fact that the number of participating chromophores increases as the concentration increases, so that more chromophores are thermally agitated. Therefore, a spatial distribution of the temperature in the sample solution is produced, which leads to a spatial variation of both the refractive index and the nonlinear absorption. As a result, phenomena such as self-defocusing, self-focusing and self-phase modulation may appear in the samples, which leads to an increase in the optical and thermal nonlinearity resulting from the 632.8 nm absorption by the samples.²² Nevertheless, the use of the optical chopper at 340 rpm, which chopped the laser beam before hitting the sample, is believed to diminish the thermal effect, leaving the

TPA process as a dominating effect in the nonlinear absorption behavior of our samples. Of course, it is needless to say that open-aperture Z-scan was carried out also on the solvent only, but no NLA was observed at the series of the laser intensities used in the experiments. It was concluded that the effect observed in the DB1/Dioxane samples was due to the solute molecule rather than the solvent.

It is worth to mention here that the nonlinear refractive index characteristics of DB1 dye were represented and discussed thoroughly in another paper²³. Briefly, the nonlinear refractive index coefficient increased with increasing the dye concentration in an exponential manner. The nonlinear refractive index coefficient values increased from -5.69×10^{-8} to -3.4×10^{-8} cm²/W as the dye concentration increased from 3.7×10^{-5} to 74.6×10^{-5} M, and were negative (Table 1); indicating that the studied solutions acted as a negative lens. Kwak's formalism²⁴ was employed in order to characterize the transmission changes and calculate the nonlinear refractive index coefficient values.

Fitting the experimental data with this theoretical approach enabled us to calculate the value of the nonlinear absorption coefficient β at each concentration. Table 1 shows the achieved results of these calculations, which clearly illustrate the expected increase in NLA coefficient values with increasing the concentration of the liquid samples.

Alsous *et al.*²⁵ studied acid blue 29/ethanol solutions, using a CW He-Ne laser at 632.8 nm, and showed that β varies as follows: 10.51×10^{-3} , 26.7×10^{-3} , and 30.49×10^{-3} cm/W at the concentrations 5×10^{-5} , 7×10^{-5} , and 8×10^{-5} M, respectively. Similarly, Mohammed Ali and Palanisamy²⁶ showed that β value of Basic Green 1 dye at the concentration of 5×10^{-5} M (using a CW He-Ne laser at 632.8 nm) was found to be 1.7×10^{-3} cm/W. These representative examples are quite comparable to ours, and are of the same magnitude, which suggests that the solutions of DB1/dioxane may have a potential application in NLO devices. Fig. 5 displays the variation nature of the increase in β values, which follows an

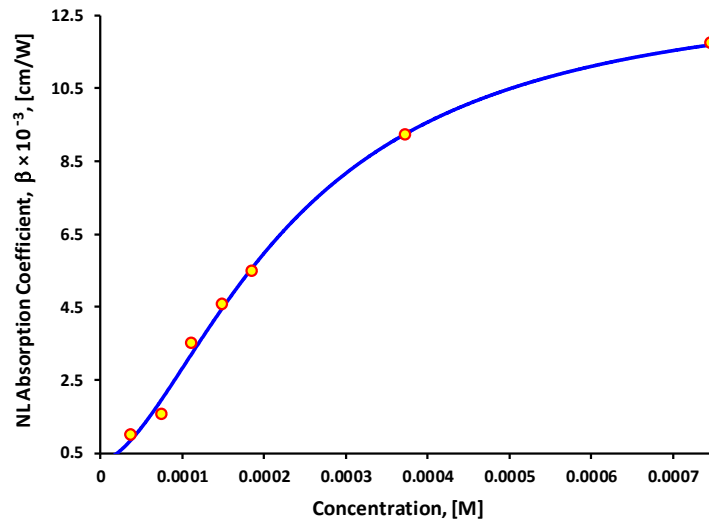


Fig. 5: Theoretical fitting of the nonlinear absorption coefficient as a function of DB1/dioxane solution concentrations.

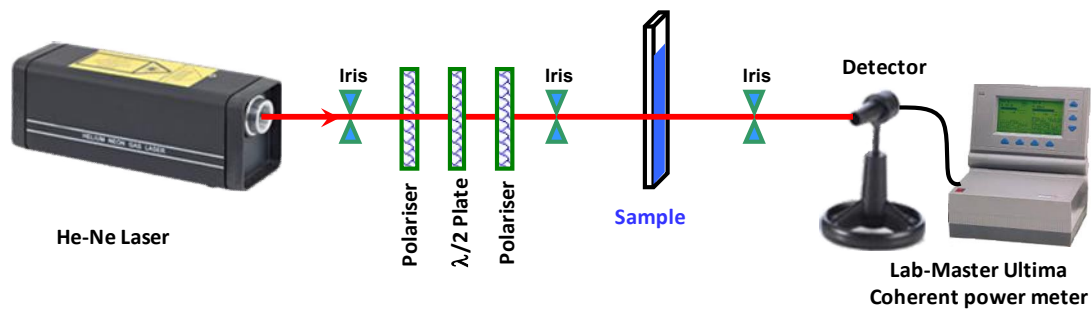


Fig. 6. The experimental set-up used for investigating the optical power limiting characteristics.

exponential fashion that can be expressed by a logistic function of the form,

$$\beta = \beta_2 + \frac{\beta_1 - \beta_2}{1 + \left(\frac{C}{C_0}\right)^p} \quad (4)$$

Where β_1 and β_2 are the initial and final values of the nonlinear absorption coefficient, respectively, at which C becomes asymptotic; C_0 is the center value of the growth section of the nonlinear absorption coefficient at which C acquires a value equal to half of the amplitude of the curve, *i.e.*, the point midway between β_1 and β_2 (the inflection point); C is the dye concentration, and p is the power of this function which controls the steepness of the curve, *i.e.*, it is the parameter that affects the slope of the area about the inflection point. This analysis showed that the absorption was nonlinear in nature and deviated quickly off the Beer-Lambert's linear behavior at as small concentration as 0.000112 M.

3.3. Optical Power Limiting Measurements

Fig.6 shows the schematic experimental apparatus used in this part of the research. An attenuator assembly (crossed Glan polarizers, and a $\lambda/2$ wave plate) was used to control the incident power on the sample, which kept the size of the laser beam unchanged as well.

No focusing devices were used, so that the beam size was as far similar as to real conditions in the lab. This procedure is useful for the fabrication of eye-safe

goggles and other protecting materials against laser radiation. Lab-Master Ultima Coherent power meter was used to carry out the measurements of the incident and transmitted power of the laser beam. However, this experimental condition was at the expense of the sample irradiance. The knife-edge technique was used to measure the laser spot size behind the sample, and it was found to be 1.24 mm of diameter. Fig. 7 shows the results of the transmission measurements in DB1/dioxane samples, at the prepared concentrations, as a function of the incident laser power. The output power varied linearly with input power at low incident powers, whereby the Beer-Lambert law was obeyed by the linear transmittance. However, at high input intensity, the sample transmission deviated away from the linearity behavior. This fashion was observed for all dye concentrations, except for the very dilute one at 3.70×10^{-5} M, which was not clear enough whether the plot has a beginning of a plateau or not due to the power limitation of our laser. The samples were absorbing the 632.8 nm line efficiently, since it resides very close to the maximum absorption wavelength of the DB1 dye. Moreover, the strong absorption was exhibited at high concentrated solutions, which indicated that the number density of dye molecules in the laser beam path was the main factor affecting the clamped level.²⁷ Fig. 7 illustrates that the optical power limiting threshold was inversely proportional to the concentration. Moreover, the data showed that as the concentration increased, a reduction in linear transmittance as well as the clamping

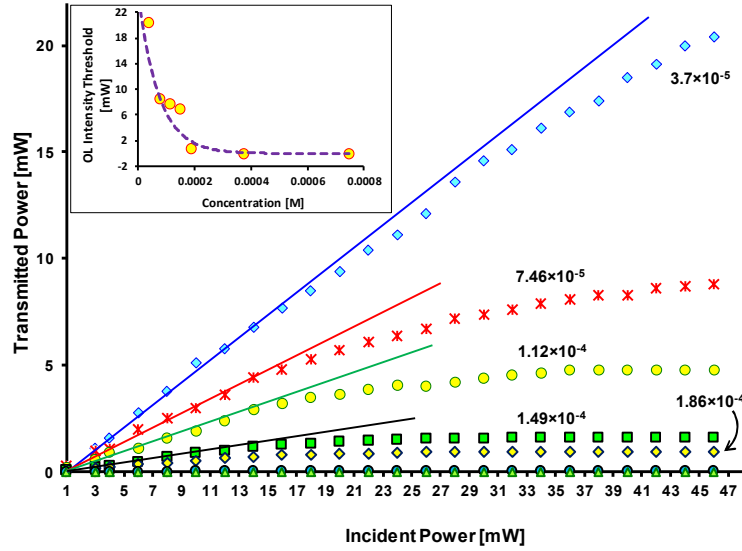


Fig. 7. Transmitted laser power as a function of the incident laser power of He-Ne laser (632.8 nm) at different dye concentrations. The inset figure shows this performance at the highest three concentrations.

level was observed. The observed results confirmed previous findings that the concentration effect is rather determinant,¹⁵ as well as confirmed strongly the exponential increase in the nonlinear absorption coefficient that has been discussed in the preceding paragraph. Moreover, the optical intensity of the limiting threshold decreases exponentially with increasing the dye concentration (the inset of Fig. 7).

A number of mechanisms have been proposed for optical power limiting, including two-photon absorption (TPA), free-carrier absorption (FCA) associated with TPA, reverse-saturable absorption (RSA) using a five-energy-level model, self-focusing, self-defocusing, thermal blooming, and nonlinear scattering.^{28,29} In a previous publication,³⁰ we have showed that both RSA, and nonlinear refraction mechanisms contribute to the power limiting behavior of DB1/dioxane samples. However, the fact that the minima in normalized transmittance (Fig. 4) existed around $z=0$, it indicates reverse saturable absorption (RSA) characteristics as explained by the five-level model.³¹ This implied also that excited-state absorption (ESA) finally enhanced the RSA process in the samples.³² This could be demonstrated by studying the cross-section values of the ground (σ_{gr}) and excited (σ_{exc}) states, of the samples, which could be calculated by,³³

$$\sigma_{gr} = \frac{\alpha}{N_a C} \quad (5)$$

$$\sigma_{exc} = \frac{4\pi h c}{\lambda^2 N_0 \tau \sigma_{gr}} \beta + \sigma_{gr} \quad (6)$$

Where N_a is Avogadro's number, h is Planck's constant, c is the speed of light in vacuum, N_0 represents the total

concentration, and τ represents the lifetime of the excited state (~ 1 ms, which is the triplet state decay time³¹). Table 1 lists the calculated values, where it shows that $\sigma_{exc} > \sigma_{gr}$ by 12 orders of magnitude, and the difference increased linearly with increasing the concentration, which confirms, once more, RSA in DB1/Dioxane samples.

4. Conclusion

We have studied the absorption performance of the organic dye disperse blue 1 in dioxane using a CW He-Ne laser at 632.8 nm as a light source. 3.70×10^{-5} , 7.46×10^{-5} , 1.12×10^{-4} , 1.49×10^{-4} , 1.86×10^{-4} , 3.73×10^{-4} , and 7.46×10^{-4} M solutions were prepared and exposed to high laser intensities. DB1 dye showed efficient nonlinear absorption characteristics upon subjecting it to elevated intensities of the laser light. The nonlinear absorption coefficient increased in an exponential manner with increasing the dye concentration. On the other hand, DB1 showed considerable optical power limiting at high laser light intensities. The observed absorption effect was at 632.8 nm, which is very close to DB1 maximum absorption, at which very dilute solution of the order of $\sim 10^{-4}$ M was sufficient to limit the transmitted power of the incident laser light by more than 90%. Thus, the investigated dye is a promising candidate for optical limiting devices under continuous wave laser at the experimental wavelength.

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