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# Broadband Nonlinearity of Trypan Blue Dye for Optoelectronic Applications

*This work presents detailed investigation on the linear absorption, emission, and nonlinear absorption and refraction of the organic dye Trypan Blue (TB) in two polar solvents water and ethanol. The nonlinear characterization was carried out by utilizing the z-scan technique with four excitation wavelengths 405, 473, 532, and 650 nm. The linear study indicates minor effects of the solvent and the major relaxation processes of the dye are non-radiative except a very small portion appears in the form of low fluorescence at the red edge of the visible region. In an intensity range (1.8-6.2 MW/m<sup>2</sup>), the TB dye does not show any nonlinear absorption properties. However, TB shows significant defocusing nonlinearity over the whole visible region. The dye shows high reliability properties of optical limiting and possibility of active optical control. The results of this study can open new insight in the TB dye usage in several photonic and optoelectronic applications.*

**Keywords:** Nonlinear optics; Optical materials; Optical switches; Trypan blue dye  
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## 1. Introduction

The great potential of laser radiation and its enormous number of applications are accompanied by its ability to harm the human eyes and sensitive optical components. This requires using protectors or optical limiters, which are devices with constant transmission up to a certain value of the input radiation power after which the output does not follow the increment of the incident power. Material with third order nonlinearity can be employed to form passive and active energy-absorbing or energy-spreading optical limiters. In addition, many other applications in the fields of photonics and electro-optics require material with specific nonlinearity in a specific spectral range. Therefore, exploring the linear and nonlinear characteristics of material has a special importance in many fields. It has been the aim of a huge number of studies, e.g. [1-5].

Organic dyes are important material in optical applications due to their unique nonlinear and spectral properties. The polarizability of dye molecules, hence its nonlinearity, is enhanced by the existence of strong delocalized  $\pi$ -conjugated electrons in the molecular system [6]. Some organic dyes have large and rapid nonlinearity besides the stability, the availability, and the ability of being incorporated in many liquid and solid hosts. The large nonlinearity is particularly required for low threshold optical limiters and the fast response is essential for the short laser pulse applications. In addition, in large nonlinear susceptibility material, relatively large nonlinear changes can be obtained in a very short propagation length. This allows the incorporating of very small nonlinear media in optical circuits.

Consequently, some organic materials have become the key components in many optical applications.

In general, material nonlinearity is determined by the nature of the material and the influence of the surroundings. For example, in liquid media, the polarity, the thermal conductivity, the hydrogen binding ability, and other solvent characteristics are proved to manipulate the nonlinearity of the solute via several mechanisms [7-10]. Furthermore, the wavelength of the excitation radiation can induce dissimilar nonlinear responses depending on the location of that wavelength relative to the absorption band of the material [11,12]. Therefore, for exploring the potential of a specific type of nonlinear material, the properties of the surroundings and the working spectral range should be taken into account.

Trypan Blue (TB) is a diazo dye that can exist in enol-azo form and keto-hydrazone form [13]. It has been widely used in Biology and optical applications, e.g. [13-16]. The linear absorption properties of TB in various solvents have been studied by Graham et al. [13]. The nonlinearity of TB at 630 nm has been used by Azarpour et al. to detect the decolorization of the dye in bacterial media [14].

In this work we comprehensively investigate the linear and nonlinear response of the organic dye Trypan Blue. Besides the linear properties, the nonlinearity of this dye was characterized in two polar solvents, water and ethanol, by using four different lasers with wavelengths spanning the whole visible range (405, 473, 532, and 650 nm). The Z-scan technique was employed to characterize the nonlinearity of the dye. In addition, employing the dye in optical limiting applications was examined for different dye concentrations in both solvents.

Furthermore, a two color setup was used to investigate the possibility of employing the TB dye in active control devices.

### 2. The theoretical background

The third order susceptibility of material ( $\chi^{(3)}$ ) consists of real ( $Re(\chi^{(3)})$ ) and imaginary ( $Im(\chi^{(3)})$ ) parts as:

$$\chi^{(3)} = \left[ \left( Re(\chi^{(3)}) \right)^2 + \left( Im(\chi^{(3)}) \right)^2 \right]^{1/2} \quad (1)$$

Both parts are related to the nonlinear refractive index  $n_2$  and the nonlinear absorption coefficient  $\beta$  as [2]:

$$Re(\chi^{(3)}) = 10^{-4} \frac{\epsilon_0 n_0^2 c^2}{\pi} n_2 \quad (2)$$

$$Im(\chi^{(3)}) = 10^{-2} \frac{\epsilon_0 n_0^2 c^2 \lambda}{4\pi^2} \beta \quad (3)$$

where  $\epsilon_0$ ,  $c$ ,  $n_0$ , and  $\lambda$  are the vacuum permittivity, speed of light, linear refractive index, and the wavelength of light.

A strong electric field of an electromagnetic wave with intensity ( $I$ ) propagating in a nonlinear medium can experience both parts of the nonlinear susceptibility. As a result of that, the refractive index ( $n$ ) and the absorption coefficient ( $\alpha$ ) will experience intensity-induced changes as [18]:

$$n(I) = n_0 + n_2 I \quad (4)$$

$$\alpha(I) = \alpha_0 + \beta I \quad (5)$$

where,  $\alpha_0$  is the linear absorption coefficient.

The nonlinear refraction can be of two opposite signs depending on the nature of material. In a sample with positive refraction the propagating light beam undergoes focusing action, while negative refraction causes defocusing. The transmittance of the sample can also change in two directions. The transmittance of material with positive nonlinear absorption coefficient decreases as the incident optical intensity increases, while the opposite happens with the negative nonlinearity.

The sign and the magnitude of the nonlinearity of low scattering media can be simultaneously obtained from the Z-scan technique [19,20]. Figure (1) depicts the working principle of this technique. Z-scan technique requires that the sample scans a focused beam through its focus. The nonlinearity of the sample leads to changes in the beam size at the far field. These changes are detected by recording the transmittance of a pinhole located at the far field (closed-aperture transmittance). The resulting relation between the normalized transmitted power and the position of the sample reveals the sign and the magnitude of  $n_2$ .

With a Gaussian beam, the difference between the highest and lowest transmittance ( $\Delta T_{p-v}$ ) can reveal the value of  $n_2$  as [20]:

$$n_2 \approx \frac{1}{0.405(1-S)^{0.25} K I_0 L_{eff}} \Delta T_{p-v} \quad (6)$$

with

$$L_{eff} = \frac{1 - e^{-\alpha_0 L}}{\alpha_0}, \quad S = \left( 1 - e^{-2r^2/w^2} \right)$$

where,  $K = 2\pi/\lambda$  is the wavenumber,  $L$  is the sample thickness,  $I_0$  is the on-axis intensity,  $r$  and  $w$  are the radii of the aperture (A1) and the beam at the aperture, respectively.

The nonlinear absorption coefficient  $\beta$  can be obtained from the variation of the optical transmittance of the sample, which is recorded by the detector D2 (open aperture transmittance), as [18]:

$$\beta = \frac{2\sqrt{2}\Delta T}{I_0 L_{eff}} \quad (7)$$

where, in the normalized open aperture transmittance curve,  $\Delta T$  is the difference between the transmittance at the focal plane and the transmittance at the linear regime.

### 3. The experimental details

The Trypan Blue dye was obtained from BDH and used with the provided quality. The two solvents used in this study, ethanol and water, were of spectroscopic purity. Solutions of the dye at concentration of 1 mM were prepared at room temperature by dissolving appropriate weight of the dye in the two solutions. Simple dilution process was used to prepare a set of different concentrations.

(CECIL CE 7200) UV-VIS Spectrophotometer was used to record the linear absorption spectra. The emission properties of the dyes were examined by measuring the laser-induced fluorescence (LIF) of the dye. In this part the dye sample is excited by 532 nm CW laser and the emitted radiation is analysed by a high resolution spectrometer (spectra-academy). A 1 mm thick quartz cuvette was used to contain the sample in the nonlinear characterization. The sample is moved in the z-scan setup shown in Fig. (1) by using a computerized stage. The open and the closed aperture transmittance were recorded simultaneously at every 15  $\mu\text{m}$  of the sample position. The nonlinearity of the samples was characterized at four wavelengths (405, 473, 532, and 650 nm) by using diode lasers. A high quality lens with 8.5 cm focal length is used to focus the laser beam.

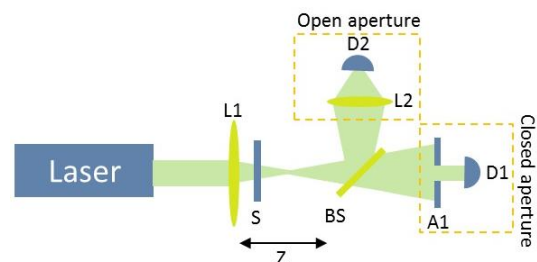


Fig. (1) A simple layout of the z-scan technique. L1: lens1, L2: lens2, S: the sample, BS: beam splitter, A: aperture, D1: detector 1, D2: detector 2

The optical limiting properties were investigated by positioning the sample directly beyond the focal plane and the transmittance was recorded as a function of the input power through an aperture of specific size. In addition, a two-colour setup, shown in Fig. (2), was used to investigate the active control

properties of the dye. In this setup a CW pump beam is chopped by a variable speed chopper and focused into the sample by using a lens with focal length of 5 cm. A He-Ne beam with very low power is used to probe the nonlinear changes in the sample. The focus of the probe was overlapped with the focus of the pump by using a charged coupled device (CCD) located at the focal plane of the lens.

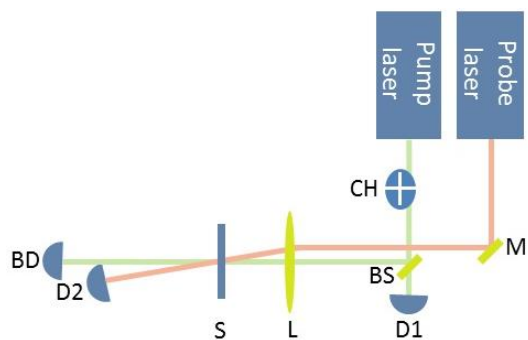


Fig. (2) A two-color setup used for investigating the possibility of active optical control, CH: chopper, M: mirror, BS: beam splitter, L: lens, S: sample, D1, D2: detectors, and BD: beam damper

4. Results and discussion

Figure (3) shows the linear absorption spectra of TB in water and ethanol. Each of the absorption spectra consists of two bands which indicate two transitions to different energy groups. The UV bands in water and ethanol are almost identical. The visible bands each consists of two merged peaks. The visible absorption band of the dye in water is wider and its peak is blue shifted by 24 nm relative to the peak in ethanol.

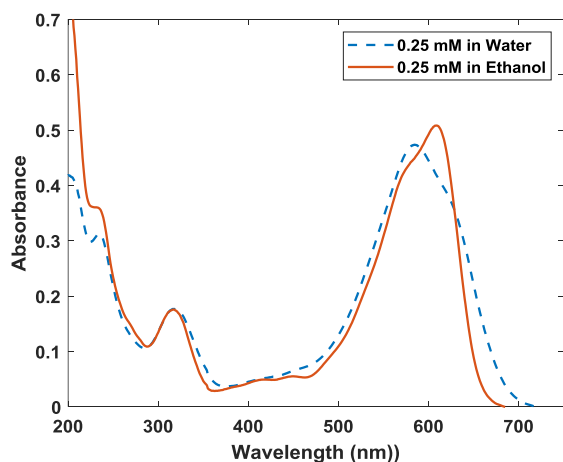


Fig. (3) The linear absorption spectra of 0.25 mM of TB in water and ethanol

The TB dye is proven to exist only in the keto-hydrazone form in water and ethanol [13]. This means that, the spectral changes in the two solvents are not due to tautomerism of the dye. Instead, these changes can be induced by the different properties of the solvents, mainly the polarity and the hydrogen binding, which can differently influence the ground and the excited energy states of the dye. However, the

oscillator strength values of the dye in the two solvents are not significantly different.

The LIF spectra of the TB dye shows extremely low fluorescence level in ethanol and zero fluorescence in water as shown in Fig. (4). At low concentration (0.01 mM) the fluorescence spectrum peaked at 656.4 nm.

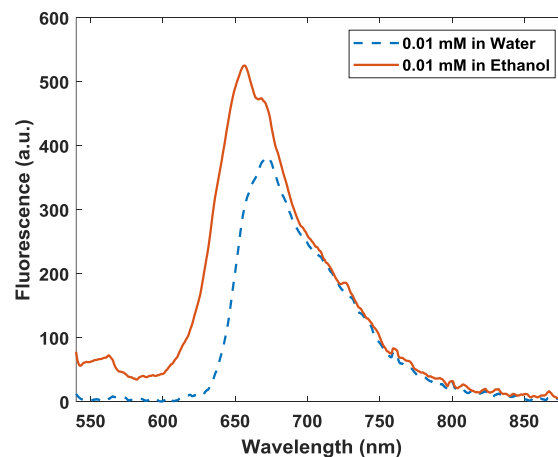


Fig. (4) The laser-induced fluorescence spectra of TB dye in ethanol

At higher concentration, 0.1 mM, the emission spectrum is quenched due to dimers formation. In addition, at high concentration, the fluorescence spectrum is red shifted due to the re-absorption of the fluorescence photons by the dye molecules themselves. The reabsorption in this case happens in the common spectral area between the absorption and the emission spectra. The low fluorescence of the TB dye reveals the domination of the non-radiative relaxation processes of this dye. This boosts the thermal nonlinearity of the dye as will be shown later.

The recorded curves by the detector D2 (open aperture curves) at all wavelengths are completely flat. This indicates that the TB dye does not show any nonlinear absorption in the range of the light intensity used in this study (1.8-6.2 MW/m<sup>2</sup>). This is true for all the tested concentrations of the dye (0.01-0.5 mM) in water and ethanol.

Figure (5) depicts the close-aperture curves of 0.25 mM TB at the four different wavelengths. The peak followed by a valley of the curves indicates that the TB dye shows defocusing refraction, with negative refractive index, in all the studied cases. The numerical values of the nonlinear refractive index are listed in table (1).

Table (1) The values of  $n_2$  of 0.25 mM TB solutions in water and ethanol

		$-n_2 \times 10^{-11} \text{ m}^2/\text{W}$			
Wavelength (nm)	Solvent	405	473	532	650
	Ethanol	5.53	38.04	43.56	18.33
	Water	1.42	4.53	4.31	5.33

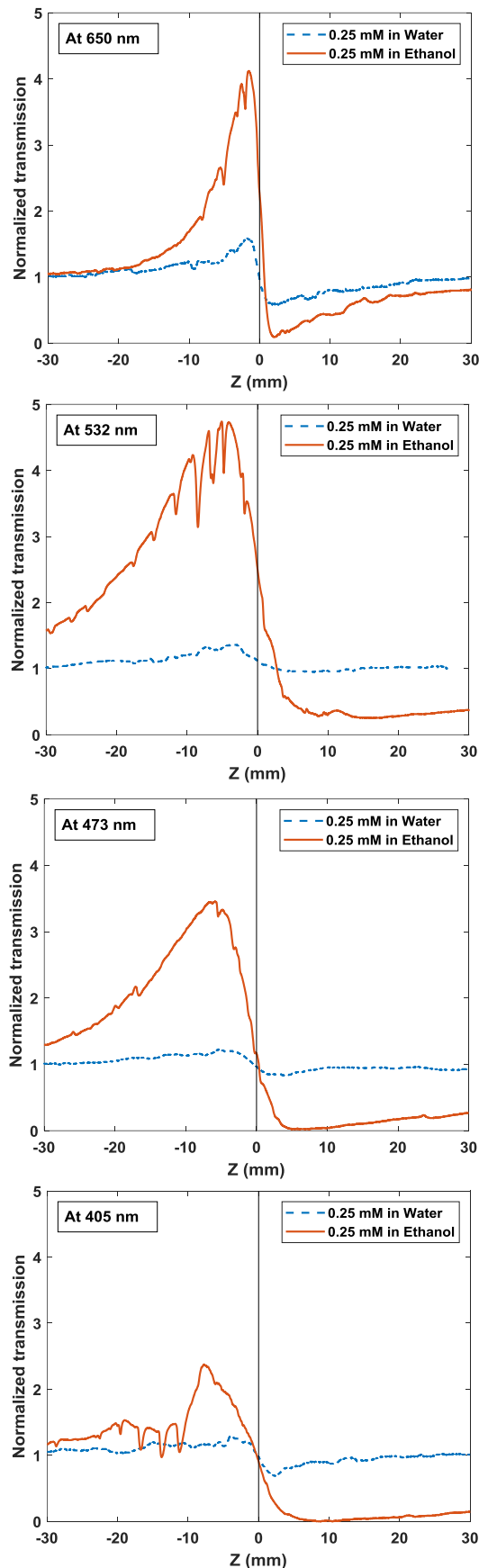


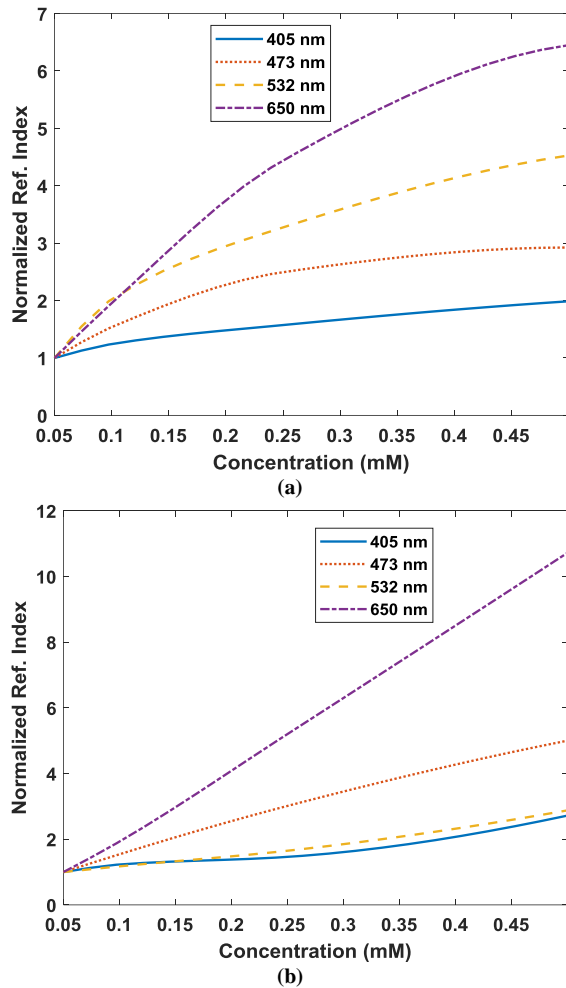
Fig. (5) The closed-aperture curves of 0.25 mM of the TB dye in water and ethanol at different excitation wavelengths

The nonlinear refraction can be via different origins, electronic perturbation, molecules arrangement or redistribution, thermal effects, and other effects [21]. The values of  $n_2$  of the TB dye solutions (in the  $10^{-6}$  cm<sup>2</sup>/W range) reveal the domination of the thermal-induced refraction changes [22]. This kind of change is mainly caused by the laser-induced temperature and density changes of the medium, which can be via thermal expansion and electrostriction effects, although the latter is not significant in highly absorbing liquids [23].

In dye solutions, some of the absorbed radiation power by the dye molecules is dissipated via non-radiative relaxation processes. This results in a local gradient of temperature centred on the axis of the beam. This gradient causes refractive index changes having spatial distribution identical to the temperature distribution. In the steady-state regime, when the excitation source is a CW, long pulsed (micro-second and longer), or high repetition rate laser, the change of the refractive index is governing by the rate of heat diffusion in the medium [24]. Therefore, in this regime, the thermal properties of the medium, the solvent in the present case, are the main parameters that determine the level of refractive index changes. This explains the large difference between the values of  $n_2$  of the aqueous solution and those in ethanol, as presented in table (1). Although, water and ethanol have different properties, in terms of polarity and hydrogen binding ability, etc., the different thermal properties of water are the main reasons of the lower  $n_2$  values of the aqueous solution of the dye in comparison with those in ethanol.

The extremely low fluorescent property of the TB dye maximizes the thermal-lensing effect, hence the nonlinearity of the solution. This is because almost the whole absorbed energy by the dye molecules is converted to heat in the solvent. The other parameter that raises the nonlinearity of the solution is the concentration of the dye, as shown in Fig. (6) which depicts the variation of  $n_2$  as a function of the dye concentration. This increase is originated from the increase of the energy absorbed by the increasing number of the dye molecule which in turn enhances the thermal-lensing effect via raising the dissipated heat into the solvent.

The curves in Fig. (6a) indicate that the growth rate of the refractive index is higher for longer excitation wavelength. In addition, the curves shows that the relationship between  $n_2$  and the concentration of the dye is not linear especially at high concentrations. This can be due to the saturation of the nonlinearity at high concentrations. The same saturation effect was observed as the intensity of the laser was increased, where the dye solutions in ethanol show lower nonlinearity at relatively higher intensities.



**Fig. (6)** The variation of the normalized nonlinear refractive index of the TB dye in a) ethanol and b) water as a function of the dye concentration at different excitation wavelengths. The values of the refractive indices are normalized to the minimum value of the lowest concentration at each wavelength

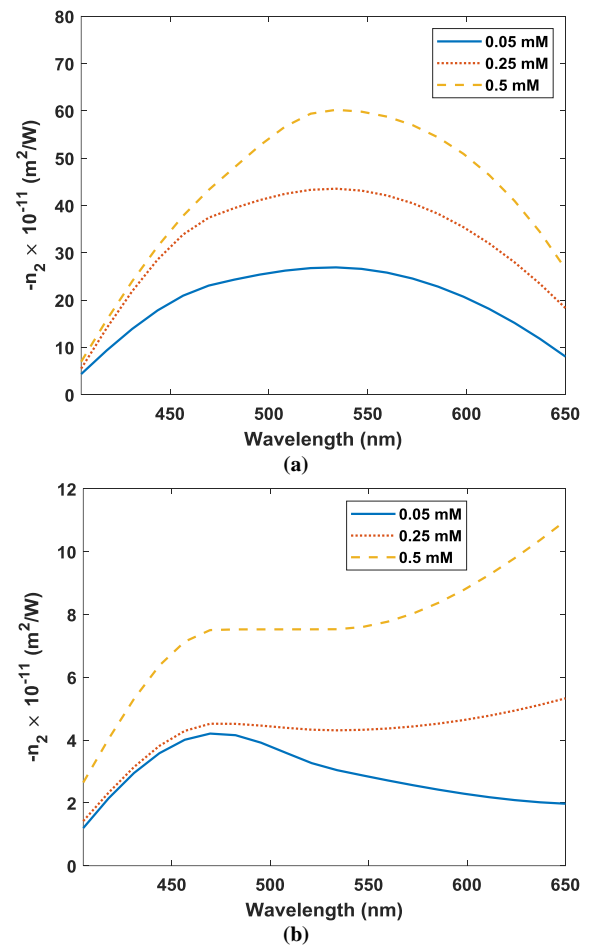
However, the value of  $n_2$  of the aqueous solution of the dye shows almost linear increase with the concentration of the dye, as shown in Fig. (6b). In addition, the nonlinearity of the dye in water increases with the intensity of the excitation laser. This behaviour is different than that in ethanol. This can indicate that the value of  $n_2$  of TB in ethanol is almost the maximum possible value of  $n_2$  of this dye. Therefore, the saturation effects happen clearly in ethanol while no saturation effect was seen in water because the values of  $n_2$  are far from the maximum values.

Furthermore, the value of  $n_2$  of the TB dye depends on the wavelength ( $\lambda$ ) of the excitation laser. The dependence of  $n_2$  on the wavelength of the incident beam is shown in Fig. (7).

In ethanol, the variation of  $n_2$  with  $\lambda$  follows the variation of the linear absorption of the dye with  $\lambda$ .  $n_2$  for all concentration peaks at 532 nm where the dye has the highest linear absorbance relative to the values at the other used wavelengths. This can indicate that, if a high resolution wavelength study is performed, e.g. by using a tuneable laser, the

maximum value of  $n_2$  could happen at the peak of the linear absorbance at 609 nm.

However, in water, Fig. (7b), the variation of  $n_2$  with the wavelength is different at different concentrations of the dye. At low concentration, the dependence of  $n_2$  on  $\lambda$  follows the dependence of the linear absorption on  $\lambda$ . However, at high concentrations,  $n_2$  continues increasing with the wavelength of the excitation light. The different behaviours at different concentrations can be due to the competition between the amounts of the absorbed energy, hence the generated heat, and the thermal conductivity of water, i.e. the speed of heat diffusion.



**Fig. (7)** The dependence of  $n_2$  of TB on the excitation wavelength in a) ethanol and b) water

The nonlinear refraction can be employed to design an energy-spreading optical limiter. This kind of limiters depends on the ability of the nonlinear sample of spreading the laser beam with defocusing power depending on the product ( $I L n_2$ ). In general, the intensity threshold ( $I_{th}$ ) and the limiting intensity ( $I_L$ ) of any energy-spreading limiter depend on the combination of the product ( $I L n_2$ ) and the ratio  $r/w$ . For a particular limiter,  $I_{th}$  and  $I_L$  increase with increasing the ratio  $r/w$  and vice versa. This feature enables adjusting the values of  $I_{th}$  and  $I_L$  of the same optical limiter to specific desired values with relatively large adjusting range. For protecting an

optical device with a fixed-size pupil, the ratio  $r/w$ , hence  $I_{th}$  and  $I_L$ , can be changed by changing the distance between the limiter and the device.

The relatively large nonlinear refraction of the TB dye enables designing a high reliability energy-spreading optical limiter with low threshold. Figure (8) presents some optical limiting curves of the TB dye. The excitation beam is at 532 nm. The curves of the dye in ethanol are recorded with  $r/w$  ratio that is two times higher than that used with the samples of the dye in water.

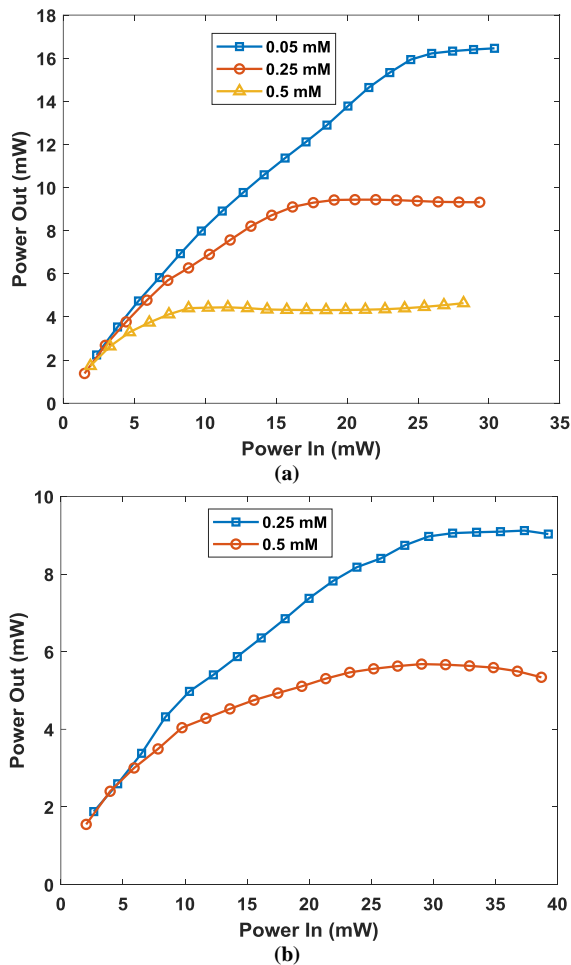


Fig. (8) The optical limiting curves of different concentrations of TB dye in (a) ethanol and (b) water at 532 nm.

As can be seen in Fig. (8a), the dye shows clear optical limiting behaviour. At low input power, the output power increases linearly with input power. However, beyond the power threshold ( $P_{th}$ ), the output power is stabilized around a certain value ( $P_L$ ). The values of  $P_{th}$  and  $P_L$ , or  $I_{th}$  and  $I_L$ , decrease with increasing the concentration of the dye. For the particular case shown in Fig. (8a), the values of  $P_{th}$  and  $P_L$  are (25.5, 16.7, and 8.4 mW) and (16.3, 9.4, and 4.5 mW) for dye concentrations (0.05, 0.25 and 0.5 mM), respectively.

The curves in Fig. (8b) reveal the optical limiting properties of TB in water. Although the lower  $r/w$  ratio, ( $P_{th}$ ) and ( $P_L$ ) of the aqueous solution are

higher than those of the dye in ethanol due to the lower values of  $n_2$  of the dye in water.

In terms of optical limiting properties, employing the dye in ethanol is preferable because the same threshold value can be obtained with lower concentration of the dye.

Using lower concentration ensures lower linear absorption therefore lower losses. The optical limiting investigation of the dye at the 532 nm wavelength represents an example of the limiting ability of TB at this wavelength. In general, TB dye can be used to design a nonlinear spreading limiter at any wavelength in the visible range, because it has the required nonlinearity over this range, as shown above. However, the optical limiting characteristics at any wavelength depend on the value of  $n_2$  at this wavelength.

In active optical control devices, the properties of the output radiation can be actively controlled by another optical source such as a laser beam. The other way of employing this concept is to detect the optical changes in a medium by a probe laser beam. In this part we show the ability of controlling the properties of a low intensity probe beam by a higher intensity pump beam via the third order nonlinearity of the TB dye.

The broadband nonlinearity of the TB dye enables employing this dye in broadband active optical devices. For examining this property, we used the setup shown in Fig. (2). Figure (9) shows an example of the readings recorded by the two detectors shown in Fig. (2). The pump in this case is a relatively high intensity beam at 405 nm and the probe is a very low intensity He-Ne beam. Similar results have been obtained when using pump beams with wavelengths (473, 532, and 650 nm).

As can be seen, the probe beam is directly derived by the pump beam. The mechanism of this control is that the probe beam is deflected by the pump-induced changes of the refractive index. The amount of this deflection depends on the value of the product ( $I_p L n_2$ ), where  $I_p$  is the intensity of the pump. In the case where the aperture of D2 is small in comparison with the lateral deflection distance of the probe, the phase difference between the pump and the probe depends on the position of the D2 relative to the direction of the probe beam. Therefore, the  $\pi$  phase difference between the two signals, shown in Figure (9), can be reversed by changing the position of D2 relative to the probe beam.

The shape of the probe intensity change, i.e. the shape of the probe pulses, depends on the build-up and the relaxation time of the nonlinearity. The nonlinearity of the dye in this case is of thermal origin, as explained above, with build-up and relaxation times in the microsecond to millisecond scale. Since this range is comparable to the duration of the pump pulse used in this work, the shape of the probe pulses changes as the repetition rate of the pump increases. At low frequencies, the probe pulses

can have some steady-state or flat regions, while at high frequencies they become like a train of triangular waves. However, with repetition rate up to 400 Hz, we found that the ratio between the maximum and the minimum of the recorded probe intensity is constant. This ensures a constant on/off ratio for any active optical control device.

The results shown in this section indicate the possibility of employing the samples of the TB dye in designing an active optical limiter or an optical switch with broadband working range.

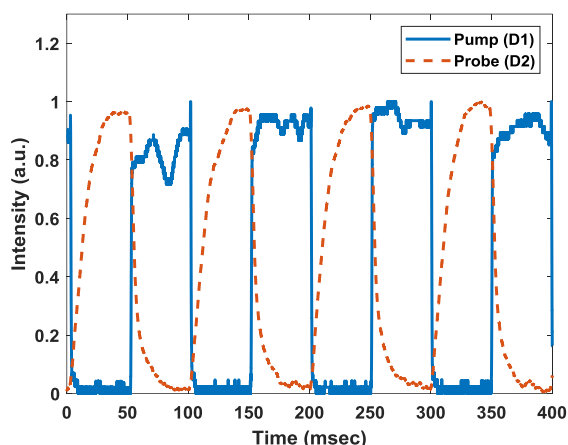


Fig. (9) The intensity of the pump and the probe beams recorded by the detectors D1 and D2 shown in Fig. (2) the nonlinear sample is a 1 mm thick quartz cell contains TB dye of concentration 0.25 mM in ethanol

## 5. Conclusion

The linear absorption of TB is slightly influenced by the characteristics of the solvent and the dye shows extremely low radiative emission in ethanol only. In the range of intensity less than  $6.2 \text{ MW/m}^2$ , the third order susceptibility of TB consists of a real part only with negative index of refraction ( $n_2$ ). This refraction depends strongly on the thermal characteristics of the solution, the concentration of the dye, and the wavelength of the excitation laser. In general, the dye shows standard characteristics of a broadband energy-spreading optical limiter. Furthermore, the study show that the TB can be employed in active optical control applications.

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