

$\chi^{(3)}$ measurements and optical limiting in Bismarck Brown Y dye

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Abstract: We investigated the third order nonlinear optical properties of Bismarck Brown Y dye. The nonlinear measurements were performed by using single beam Z-scan technique with cw solid state laser at 473 nm. The nonlinear absorption coefficient is calculated using the open aperture Z-scan data, while its nonlinear refractive index is measured using the closed aperture Z-scan data. The nonlinear refractive index and absorption coefficient are found to be in the order of $10^{-7} \text{ cm}^2/\text{Watt}$, 10^{-3} cm/Watt , respectively. The third order nonlinearity $\chi^{(3)}$ is measured using Z-scan data. The optical limiting behavior is investigated by measuring the transmission of the sample. The results indicate that Bismarck Brown Y is a potential candidate for low-power optical limiting application.

Keywords: Optical limiting; nonlinear refraction index; Z-scan; cw laser.

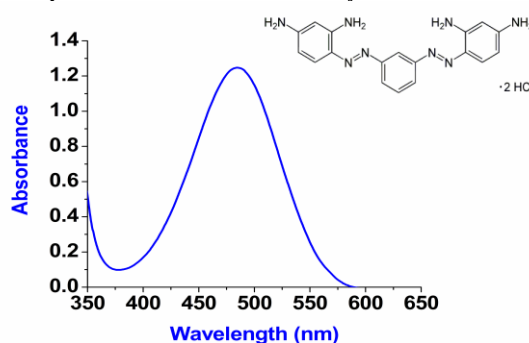
I. Introduction

The nonlinear optical properties of materials can be used to control the phase, the state of polarization, or the frequency of light beams. With the emergence of photonic technologies in areas such as telecommunications where information is coded, transported, and routed optically, there is a strong technological demand for high-performance NLO materials. Organic molecules are promising candidates for these nonlinear optical applications [1-5]. A wide variety of materials have been investigated for third-order nonlinear optics, among which organic materials are attractive because of their optical and electronic properties which can be tuned and tailored by structural modification. The third-order optical nonlinearity includes optical bleaching (i.e., saturation) or reverse saturation in the absorption aspect, whereas self-focusing or self-defocusing occurs in the refraction aspect. Of the various techniques available, Z-scan method [6,7] is a simple and effective tool for determining nonlinear properties and is used widely in material characterization because, it provides not only the magnitudes of the real and imaginary parts of the nonlinear susceptibility, but also the sign of the real part. In this paper, we report on our experimental investigation of the third order nonlinear optical susceptibility $\chi^{(3)}$ in Bismarck Brown Y the optical nonlinearity induced in dye Bismarck Brown Y by cw diode laser with an output power of 4.5 mW at 473 nm was studied using Z-scan technique, based on the sample-induced changes in beam profile at the far field. The optical limiting behavior of the dye has been studied.

II. Experimental

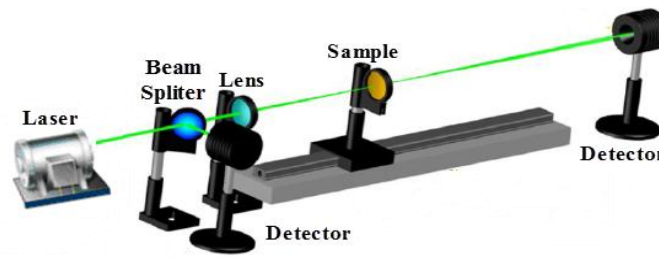
The molecular structure of the Bismarck Brown Y dye and the linear absorption spectrum of the dye dissolved in double distilled water at 3mM concentrations is shown in Fig. 1, which was acquired using a UV-VIS spectrophotometer (Type: CECEL 3500).

Figure 1 UV-Vis absorption spectrum of Bismarck Brown Y dye. Inset shows the chemical structure of dye



The schematic diagram of Z-scan technique is as shown in Fig. 2. By properly monitoring the transmittance change through a small aperture at the far field position (closed aperture), one is able to determine the amplitude of the phase shift. By moving the sample through the focus without placing an aperture at the detector (open aperture) one can measure the intensity dependent absorption of the sample. When both the methods (open and closed) are used in measurements of the ratio of signals determines the nonlinear refraction of the sample.

Figure 2 Z - scan experimental setup.



II. Nonlinear optical measurement

The nonlinear coefficients of the Bismarck Brown Y was measured by using Z-scan technique, which is a well-known technique that allows the simultaneous measurement of both nonlinear absorption coefficient β and the nonlinear refractive coefficient n_2 . A closed aperture scheme allowed us to determine the value of the nonlinear refractive index of the sample and an open aperture scheme is applied in order to determine the value of the nonlinear absorption coefficient β . Fig. 3 show the closed aperture Z-scan for solution of Bismarck Brown Y in double distilled water at 3 mM. The peak followed by a valley-normalized transmittance obtained from the closed aperture Z-scan data, indicates that the sign of the refraction nonlinearity is negative i.e. self-defocusing. The defocusing effect shown in Fig. 3 is attributed to a thermal nonlinearity resulting from absorption of radiation at 437 nm. Localized absorption of a tightly focused beam propagating through an absorbing dye medium produces a spatial distribution of temperature in the dye solution and consequently, a spatial variation of the refractive index, that acts as a thermal lens resulting in phase distortion of the propagating beam.

The difference between normalized peak and valley transmittance ΔT_{p-v} can be measured by Z-scan technique.

The peak to valley ΔT_{p-v} is linearly related to the on- axis phase distortion $\Delta\Phi_0$ of the radiation passed through the sample [8, 9] the relation is defined as,

$$\Delta T_{p-v} = 0.406(1-S)^{0.25}|\Delta\Phi_0| \quad \dots\dots\dots (1)$$

and

$$\Delta\Phi_0 = kn_2I_0L_{eff} \quad \dots\dots\dots (2)$$

Where $S = 1 - \exp(-2r_a^2/\omega_a^2)$ is the aperture linear transmittance with r_a is the aperture radius and ω_a is the beam radius at the aperture in the linear region, I_0 is the intensity of the laser beam at focus $z = 0$, $L_{eff} = (1 - \exp(-\alpha_0 L))/\alpha_0$ is the effective thickness of the sample, L is the thickness of the sample, α_0 is linear absorption coefficient of the Bismarck Brown Y dye solution and $k = 2\pi/\lambda$ is the wave number. The nonlinear refractive index n_2 can be obtained from Equations 1 and 2, and the corresponding change in the refractive index $\Delta n = n_2 I_0$.

In the Z-scan measurement, the effective thickness of the samples is 0.034 mm, the linear transmittance of the aperture was $S=0.61$ and the optical intensity at the focus point is 5.81 kWatt/cm^2 .

The nonlinear absorption coefficient β (cm/W) can be calculated using the Eq. [10].

$$\beta = \frac{2\sqrt{2}\Delta T}{I_0 L_{eff}} \quad \dots\dots\dots (3)$$

where ΔT is the normalized transmittance for the open aperture.

Figure 3 Closed aperture Z-scan data for Bismarck Brown Y.

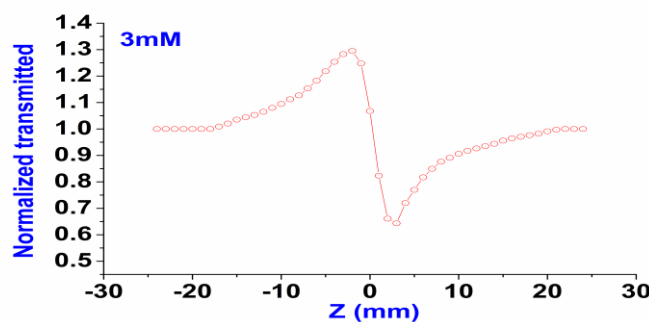
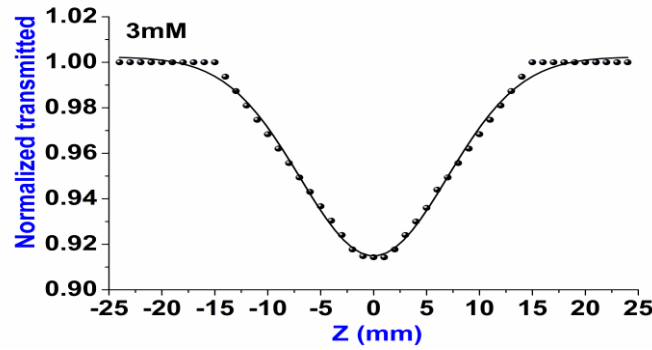


Fig.4 shows the open aperture (S=1) Z-scan curve of sample. The enhanced transmission near the focus is indicative the for TPA process at high intensity ($z = 0$).

Figure 4 Open aperture Z-scan data for Bismarck Brown Y.

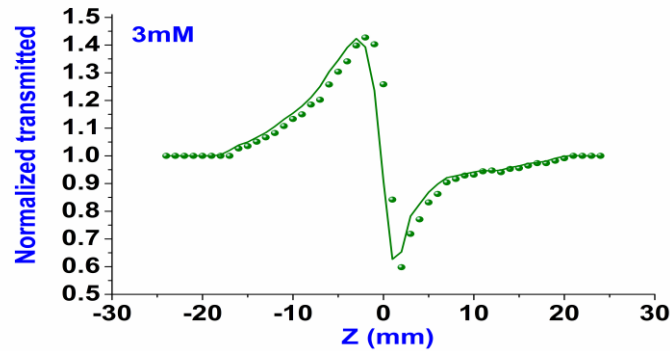


In order to extract the pure nonlinear refraction part, we have computed the value of the closed aperture (CA) data by the open aperture (OA) data. Fig.5 shows the ratios of the Z-scan data for the dye dissolved in double distilled water at 3 mM concentration. Table 1 summarizes the values of nonlinear refractive indices, nonlinear absorption coefficients, and third-order susceptibilities of the dyes. The experiment was repeated for the pure solvent (double distilled water) to account for its contribution, but no significant measurable signals were produced in either the open or the closed Z-scan traces.

Generally the measurements of the normalized transmittance versus sample position, for the cases of closed and open aperture, allow determination of the nonlinear refractive index, n_2 , and the reversible saturation absorption (RSA) nonlinear coefficient, β [11].

Here, since the closed aperture transmittance is affected by the nonlinear refraction and absorption, the determination of n_2 is less straight forward from the closed aperture scans. Therefore, it is necessary to separate the effect of nonlinear refraction from that of the nonlinear absorption.

Figure 5 pure Z-scan data for Bismarck Brown Y dye.



The third-order nonlinear optical susceptibility $\chi^{(3)}$ was determined with the Z-scan technique in a single beam configuration. The laser beam is focused onto the sample and the total power of the transmitted beam, as well as the power in its central part, is recorded in the far field as a function of the sample position along the beam axis z . The experimental measurements of n_2 , and β allow one to determine the real and imaginary parts of the third-order nonlinear optical susceptibility $\chi^{(3)}$ according to the following relations [12-14],

$$\text{Re } \chi^{(3)} (\text{esu}) = \frac{10^{-4} \epsilon_0 c^2 n_0^2 n_2 (\text{cm}^2 / \text{W})}{\pi} \dots\dots\dots (4)$$

$$\text{Im } \chi^{(3)} (\text{esu}) = \frac{10^{-2} \epsilon_0 c^2 n_0^2 \lambda \beta (\text{cm} / \text{W})}{4\pi^2} \dots\dots\dots (5)$$

where ϵ_0 is the vacuum permittivity and c is the light velocity in vacuum.

According to Eqs. 4 and 5, these values of n_2 and β can be used to calculate $\chi^{(3)}$. The absolute value of $\chi^{(3)}$ for the Bismarck Brown Y solution was calculated from $|\chi^{(3)}| = [(\text{Re}(\chi^{(3)}))^2 + (\text{Im}(\chi^{(3)}))^2]^{1/2}$, and it comes out to as shown in Table 1. It can be observed that the value of $\chi^{(3)}$ for the dye in double distilled water increases with concentration increases.

It is worth noting that the value of $\chi^{(3)}$ for the Bismarck Brown Y studied is larger than those of some representative third-order nonlinear optical materials such as organic polymers and organic metal [15–17] suggesting that the aqueous solution of Bismarck Brown Y may have a potential application in nonlinear optical devices.

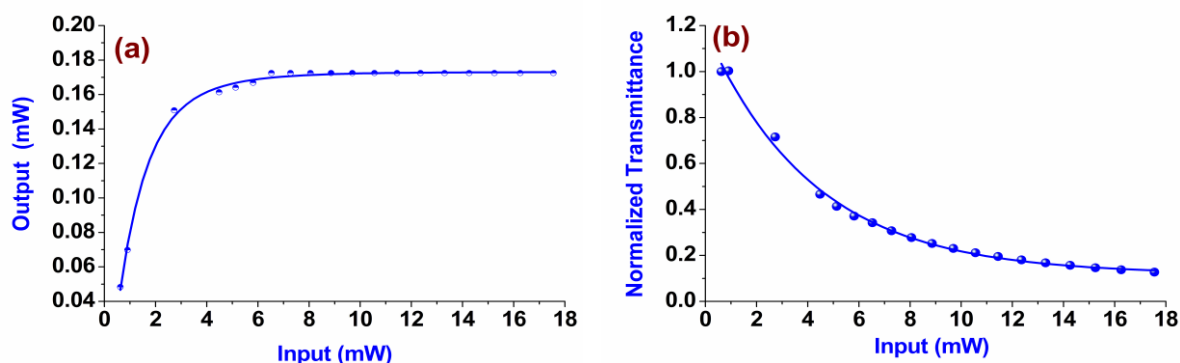
Table1 Nonlinear optical coefficients for aqueous solution of dye

Dye mM	$n_2 \times 10^{-7} \text{ cm}^2/\text{W}$	$\beta \times 10^{-3} \text{ cm/W}$	$\text{Re } \chi^{(3)} \times 10^{-6}(\text{esu})$	$\text{Im } \chi^{(3)} \times 10^{-6}(\text{esu})$	$ \chi^{(3)} \times 10^{-6}(\text{esu})$
3	9.85	12.21	1.25	5.84	5.97

III. Optical limiting

To investigate the optical limiting activity of the Bismarck Brown Y, the nonlinear transmissions were measured at 3 mM concentration. The variation of output power with input power is shown in Fig. 6(a), where an instantaneous response to the incident light is observed. Fig. 6(a) shows the clear optical limiting behaviour of the sample solution. As can be seen, the output in a sample increases with increasing incident power up to a limiting threshold, where the output power is limited. The optical limiting threshold determines the ability of the limiter and it is noted that the lower the threshold value, the better the optical limiter. Fig. 6(b) show the normalized transmission curves as a function of the incident input power for different concentrations of sample solution. The optical limiting thresholds (defined as the incident input power where the transmission reduces by 50%) are approximately 4.27 mW.

Figure 6 (a) Optical limiting at 3mM concentration, (b) Normalized transmission of optical limiting.



The input power is in the range of (0–18) mW. As is shown clearly for incident beam power of above 4.5 mW, the transmission becomes nonlinear. At incident power above 7 mW the output power tends to be constant, because its nonlinear absorption coefficient increases with increase in the incident irradiance. Generally in liquids, where the thermal expansion is large, high absorbance of the nonlinear material at the corresponding wavelength leads to increase in the temperature and density of the sample. Heating due to laser absorption is the responsible mechanism for changing the absorption coefficient and optical limiting effect [18–20]. Results confirm that Bismarck Brown Y solution is a good candidate for optical limiting at 473 nm cw laser.

V. Conclusion

The nonlinear refraction index n_2 and nonlinear absorption coefficient β , of Bismarck Brown Y solution was studied using a single beam Z-scan technique under cw laser with excitation at 473 nm. The Z-scan measurements indicated that the dye exhibited large nonlinear optical properties. We have shown that the nonlinear absorption can be attributed to a saturation absorption process, while the nonlinear refraction leads to self-defocusing in this dye. The optical nonlinearity of the dye may be due to laser heating induced nonlinear effect. A laser beam, while passing through an absorbing media, induces temperature and density gradients that change the refractive index profile. This intensity-induced localized change in the refractive index results in a lensing effect on the optical beam. It is found that the observed nonlinear absorption is caused by a reversible saturation absorption process. $\chi^{(3)}$ is calculated for different cases. These results are quite encouraging for possible applications in nonlinear optical devices. Based on nonlinear refraction the sample be haved as good optical limiters at low incident power up to 7mW, indicating this sample find potential applications in optical limiting and signal processing applications.

VI. Reference

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