Third-order nonlinearities and optical limiting properties of rose Bengal at 532 nm wavelength

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ISSN –1817 –2695

(Received 9/1/2007 , Accepted 25/4/2007)

Abstract

The nonlinear refractive index n_2 and nonlinear absorption coefficient β for aqueous solution of rose Bengal were obtained using closed aperture and open aperture Z-scan technique at the same wavelength. The nonlinear refractive index n_2 shows negative nonlinearity of the sample and it is measured to be $n_2 = -5.6 \times 10^{-8}$ cm^2 /W. Moreover, a variation of nonlinear refractive index with intensity is observed. The nonlinear absorption coefficient is measured to be $\beta = -8.3 \times 10^{-3}$ cm/W. Also the optical limiting property of the sample has been investigated using frequency doubled Nd:YAG laser.

Key words: Nonlinear optics, Nonlinear refractive index, Z-scan technique, Optical limiting, Dye.

1. Introduction

With the extensive use of continuous wave (cw) lasers at power levels ranging from μW to kW in various applications, the need for protecting the human eye and the sensors used in handling the cw output has become increasingly important. Under cw illumination, the form of optical nonlinearity exhibited by materials is predominantly refractive rather than absorptive [1,2] and suitable schemes based on nonlinear refraction have to be exploited for obtaining the limiting action. Certain materials such as liquid crystals, prophyrins, organics such as azobenzene, etc, are known to be optically nonlinear under cw laser illumination [3-5]. The refractive index of these materials depends on the input intensity, resulting in either focusing or defocusing effects on the incident laser beams. The defocusing effect under cw laser irradiation, usually associated with nonlinearity of thermo-optics origin, can be used for the design of an optical limiting devices. An aperture can be used to limit the cross section of a beam defocusing by the nonlinear medium, thereby controlling the output intensity. This type of optical limiting based on the nonlinear refraction has been demonstrated successfully with a few materials such as pthalocyanines and InO_2 [6-8].

Xanthene dyes were identified and characterized as materials suitable for optical phase conjugation (OPC) by degenerating four wave mixing in solid matrices of gelatin, poly vinyl alcohol (PVA) and boric acid glass [9,10], high density optical data storage [11] and nonlinear optics [12]. The present paper reports the study of nonlinear refractive index and nonlinear absorption coefficient of aqueous solution of an xanthene dye, namely rose Bengal by the Z-scan technique using cw diode pumped Nd:YAG laser (frequency doubled) of total power 50 mW at 532 nm. Also the optical limiting property of this sample has been studied. We found that the dye has large nonlinear refractive index and nonlinear absorption coefficient. The negative nonlinear refractive index and saturation absorption coefficient were measured for this dye.

2. Experiment

Z-scan technique has been widely used to study optical nonlinear properties of various materials [13,25]. It is a simple and sensitive technique in determining the effective nonlinear coefficient n_2 of cubic nonlinear optical materials. A Gaussian beam is focused by a positive lens. An aperture and a photodetector are centered at the axis of the lens and are placed at the far field. The Gaussian beam through the sample will defocus for negative nonlinear materials or will focus on positive nonlinear materials. So the power through the aperture will be changed with the distance between the sample and the waist of the focused beam. The transmittance through the aperture as a function of the distance between the sample and the beam waist is called Z-scan curve. A pre-focus peak followed by a post-focus valley indicates that the nonlinear property of the sample is negative and the lensing effect is defocusing; a pre-focus valley followed by a post-focus peak means that the nonlinear property of the sample is positive and the lensing effect is focusing.

The UV-VIS (Ultraviolet-Visible) absorption spectrum of the dye was recorded using an UV-VIS spectrophotometer (UV-2401 PC SHIMAZU). The optical absorption of the rose Bengal dye in water with 0.01 mM concentration shows an absorption peak (λ_{max}) at 545 nm as shown in figure 1.

The Z-scan experiments were performed using a 532 nm diode pumped Nd:YAG laser beam (Coherent, Compass 215M-50), which was focused by 3.5 cm focal length lens. The laser beam waist ω_o at the focus was measured to be 14.5 μm and the Rayleigh length Z_R = 1.24 mm. A 1 mm wide optical cell containing the aqueous solution of rose Bengal was translated across the focal region along the axial direction, that is the direction of the propagation of the laser beam. The transmission of the beam through an aperture placed in the far field was measured using photodetector fed to the digital power meter (Field master Gs-coherent). For an open aperture Z-scan, a lens replaced the aperture to collect the entire laser beam transmitted through the sample.

The experimental set-up for the demonstration of optical limiting of the laser beam by rose Bengal under cw laser illumination is shown in figure 2. This is similar to the standard Z-scan geometry and the same laser was used as for the Z-scan experimental set-up. Additionally a varying beam splitter was used to vary the input power. The cuvette containing the nonlinear medium was placed just after the focal point. An aperture A is used to control the cross-section of the beam coming out of the sample cuvette. This beam is then made to fall on the optical sensor, a photodectector in this case. The input laser power is varied systematically and the corresponding output power values were measured by the photodector.

3. Results and discussion

3.1 Nonlinear optical properties

The typical closed-aperture Z-scan curve of the 0.4 mM concentration of aqueous solution of the rose Bengal at I_o = 4.5 kW/cm^2 incident intensity, indicating the normalized transmittance, is shown in figure 3a. The curve is characterized by a pre-focal peak followed by a post-focal valley, which implies that the nonlinear refractive index of aqueous solution of rose Bengal is negative ($n_2 < 0$). Figure 3b shows the typical Z-scan data for the open aperture setup. The enhanced transmission near the focus is indicative of the saturation of absorption at high intensity. Absorption saturation in the sample enhances the peak and decreases the valley in the closed aperture Z-scan, thus distorting the symmetry of the Z-scan curve about z =0. The defocusing effect shown in figure 3a is attributed to a thermal nonlinearity resulting from absorption of radiation at 532 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 nonlinear absorption coefficient β (cm/W) can be calculated using the equation [27].

$$\Delta T = \frac{\beta I_o L}{2\sqrt{2}} \qquad (1)$$

where ΔT is the normalized transmittance for the open aperture, L is the thickness of the sample and I_o is the intensity of the laser beam at focus. The nonlinear absorption coefficient β (cm/W) is calculated from the open aperture normalized transmittance in figure 3b. The obtained β of the aqueous solution of roes Bengal is -8.3 x 10^{-3} cm/W.

The nonlinear refractive index n_2 is estimated by the following equation

$$n_2 = \frac{\lambda \Delta T_{p-v}}{0.812\pi(1-S)^{0.25} L_{eff} I_O} \quad (2)$$

where $L_{eff} = [1-\exp(-\alpha L)]/\alpha$ is the effective thickness of the sample, α is the linear absorption coefficient, λ is the laser wavelength, S is the aperture linear transmittance and ΔT_{p-v} is the difference between the normalized peak and valley transmittances for the closed aperture. In order to obtain quantitatively the nonlinear refractive index, a simple and approximate method [27] was used, in which the closed aperture data were divided by the corresponding open aperture data, and results were shown in figure 3c. The calculated n_2 of the aqueous solution of rose Bengal is -5.6 x 10^{-8} cm^2/W.

Variation of the nonlinear refractive index with respect to the incident beam intensity for 0.4 mM concentration of aqueous solution of rose Bengal is shown in figure 4. It is found that the third order nonlinear refractive index is linearly dependent on the incident intensity. These results suggest that the aqueous solution of rose Bengal may have a potential application in nonlinear optical devices.

3.2 Optical limiting

Optical limiting occurs when the optical transmission of a material saturates with increasing laser intensity, a property that is desirable for the protection of sensors and human eyes from the intense laser radiation. The optical power limiting property of the aqueous solution of rose Bengal is measured with the same laser used in section 2. Figure 5 gives the optical limiting characteristics at room temperature for the sample. The sample shows very good optical limiting behaviour arising from nonlinear refraction. The output power rises initially with the increase in input power, but after a certain threshold value, the sample starts defocusing the beam resulting in a greater part of the beam cross-section being cut off by the aperture. Thus the transmittance recorded by the photodetector remained reasonably constant showing a plateau region. The threshold power value at which the limiting occurs is found to be 1.5 mW. The experiment is carried out with solvent alone afforded no detectable optical limiting effect. All these indicate that solvent contribution is negligible.

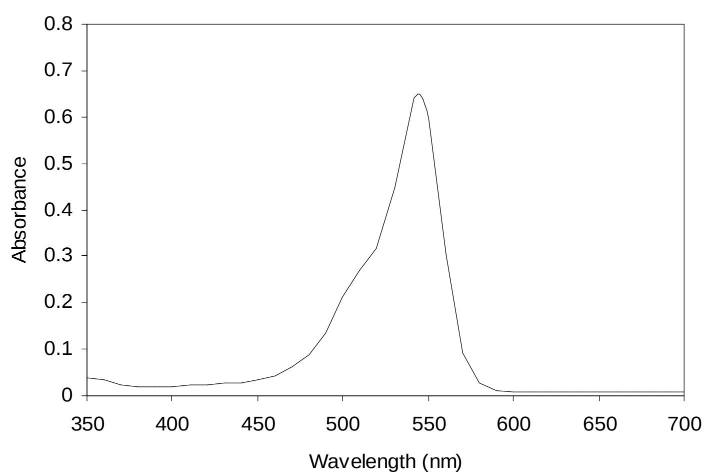

Fig.1 UV-VIS absorption spectrum of rose Bengal dye.

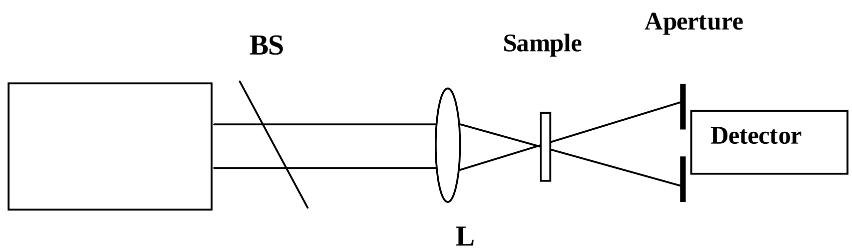

Fig.2 Experimental set-up for the demonstration of optical limiting in rose Bengal. L is the lens and BS is the varying beam splitters.

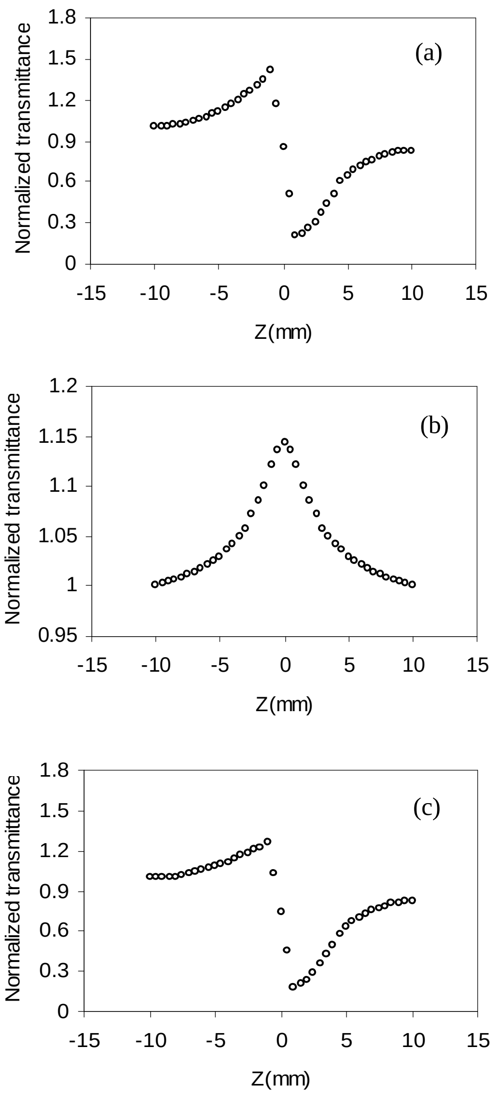

Fig.3 Z-scan data of the 0.4 mM concentration of aqueous solution of rose Bengal at I_o = 4.5 kw/cm^2 (a) closed aperture scan (b) open aperture scan and (c) the division of a by b.

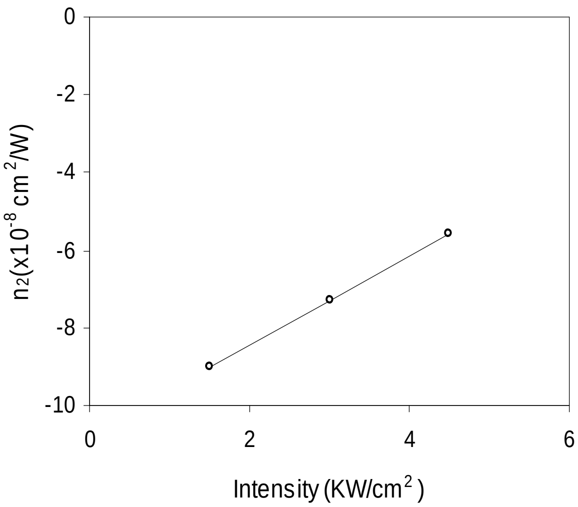

Fig. 4 Variation of n_2 with intensity for 0.4 mM concentration of aqueous solution of rose Bengal.

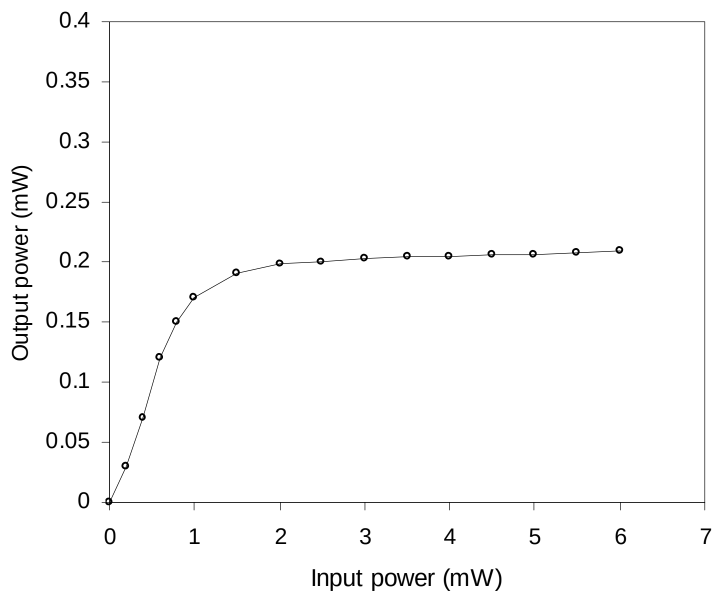

Fig 5. Output power as a function of input power for 0.4 mM concentration of aqueous solution of rose Bengal using closed aperture.

4. Conclusion

The nonlinear refraction as a function of intensity was observed for aqueous solution of rose Bengal in a Z-scan experiment with 532 nm diode pumped Nd:YAG laser. The Z-scan measurements indicated that the dye exhibited large nonlinear optical properties. For 0.4 mM concentration of the dye solution, the n_2 and β values were found to be -5.6×10^{-8} cm^2 /W and -8.3×10^{-3} cm/W, respectively. The measured absorption curves indicate that the nonlinear absorption is a saturation absorption process. Also the aqueous solution of rose Bengal have been observed as a very interesting as optical limiting material.

Acknowledgements

I would like to thank professor P. K. Palanisamy of the University of Anna, Department of Physics, India for using his lab to do this work.

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حساب معامل الانكسار والامتصاص اللاخطي لصبغه روس بنغال بأستخدام تقنيه المسح بأتجاه Z

قصي محمد علي حسن

الخلاصه

في هذا البحث تم حساب معامل الانكسار والامتصاص اللاخطي لمحلول مائي لصبغه روس بنغال بأستخدام تقنيه المسح بأتجاه Z للفتحه المغلقه والمفتوحه ولنفس الطول الموجي . لقد وجد ان معامل الانكسار اللاخطي لهذه الصبغه يظهر صفات لاخطيه سالبه ووجد انه يساوي n_2=-5.6 x 10^{-8} cm^2/W . وانه يتغير مع تغير الشده. معامل الامتصاص اللاخطي وجد انه يساوي β= -8.3 x 10^{-3} cm/W . كذلك تم دراسه الخواص الحدوديه للصبغه بأستخدام ليزر Nd:YAG .