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The Study of the Nonlinear Optical Properties of Solutions under CW Laser Illumination

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Abstract

The nonlinear optical properties of janus green in tetrahydrofuran solvent (THF) were studied using single beam Z-scan technique with a continuous-wave laser radiation at 635 nm of an output power of 40 mW. All the solution samples showed large nonlinear refractive indexes and absorption coefficient of the order of 10^{-7} cm^2/W and 10^{-3} cm/W, respectively. The concentration-dependent nonlinear refractive index was also investigated. The solvent of spectroscopic grade and was used without further purification. All the solutions used for the study were freshly prepared.

Keywords*:* nonlinear refraction index , Z-scan, azo dye, cw laser.

1. Introduction

Azo dyes have many advantages over other nonlinear optics (NO) materials. The photoisomerization of azo molecules it modifies the linear and nonlinear polarizability of them as well as optical nonlinear refraction. Since the optical properties of azo molecules can be controlled optically, they have intrigued considerable interest of people [1,2]. The nonlinear optical phenomena of azo dyes can result from electronic response and/or nonelectronic one. The electronic nonlinearity is induced by either population redistribution or distortion of electronic clouds. A molecule undergoes a transition from its ground state to its excitation state after absorbing a photon. The dipole moment of the molecule changes during such a transition. The change in the dipole moment will give birth to electronic nonlinearity. A nonelectronic response is a non-radiative interaction such as cis-trans isomerism, the changes in density and temperature [3–5]. It has been well known that the nonlinear optical behavior of materials can vary greatly by changing different laser duration or different laser wavelengths. Thus, studies about the mechanism of their nonlinear optical response with different laser duration or different laser wavelengths are expected to be more interesting and important. If the nonlinear mechanism is understood for certain laser pulses, the NO properties optimization can be well accomplished. Z-scan technique is a simple and effective tool to determine the nonlinear properties [6]. It has been widely used in material characterization because it provides not only the magnitudes of the real part and imaginary part of the nonlinear susceptibility, but also the sign of the real part. Both nonlinear refraction and nonlinear absorption in solid and liquid samples can be measured easily by Z-scan technique, which use the change of transmittance of nonlinear materials [5].

In this work, we demonstrate the optical nonlinearities of a janus green dye in (THF) through Z-scan technique under laser excitation at 635 nm cw solid state laser with an output power of 40 mW.

2. Experimental

The solution samples of the azo dye were prepared in tetrahydrofuran (THF). The sample was contained in a 1mm quartz curette, and have a refractive index of 1.33. The molecular structure of the janus green is shown in Figure1.The linear absorption spectrum of the azo dye solution with concentration of 0.07mM in tetrahydrofuran (THF) solvent is shown in Figure 2, using a UV–VISNIR spectrophotometer (Type:CECIL –CE-3550) .

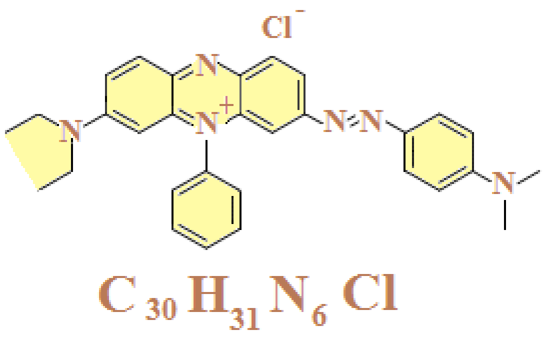


Figure 1 Chemical structure and molecular formula of janus green dye.

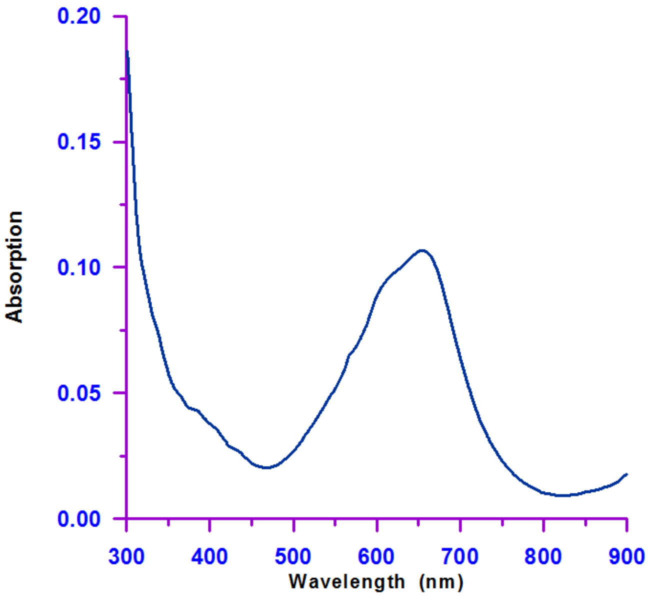


Figure 2 UV-VIS absorption spectrum of janus green dye in tetrahydrofuran solvent.

The Z-scan experiments were performed using a 635 nm solid state laser beam, which was focused by 50 mm focal length lens. The laser beam waist ω_0 at the focus is measured to be 19.36 mm and the Rayleigh length measured Z_R =1.85 mm. The schematic of the experimental set up used is shown in Figure 3. A 1mm wide optical cell containing the solution of janus green is translated across the focal region along the axial direction that is the direction of the propagation laser beam. The transmission of the beam through an aperture placed in the far field was measured using a photodetector fed to the digital power meter. For an open aperture Z-scan, a lens was used to collect the entire laser beam transmitted through the sample replaced the aperture.

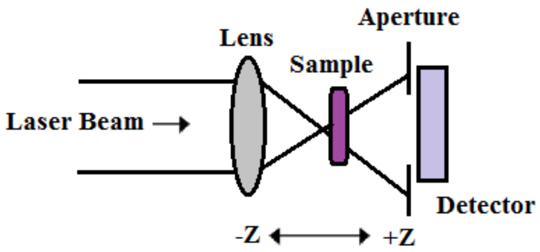


Figure 3 Schematic diagram of experimental arrangement for the Z-scan measurement.

3. Results and Discussions

The third-order nonlinear refractive index ,n_2, and the nonlinear absorption coefficient, β, of the azo dye in tetrahydrofuran at various concentrations for the incident intensity I_0 = 6.792 kW/cm^2 were evaluated by the measurements of Z-scan. Figure 4 shows the open Z-scan curve for the dye in solution at various concentrations. The typical Z-scan data with fully open aperture is insensitive to nonlinear refraction; therefore, the data is expected to be symmetric with respect to the focus, but the absorption saturation in the sample enhances the peak and decreases the valley in the closed aperture Z-scan curve and results in the distortions in the symmetry of the Z-scan curve about Z = 0 [7].

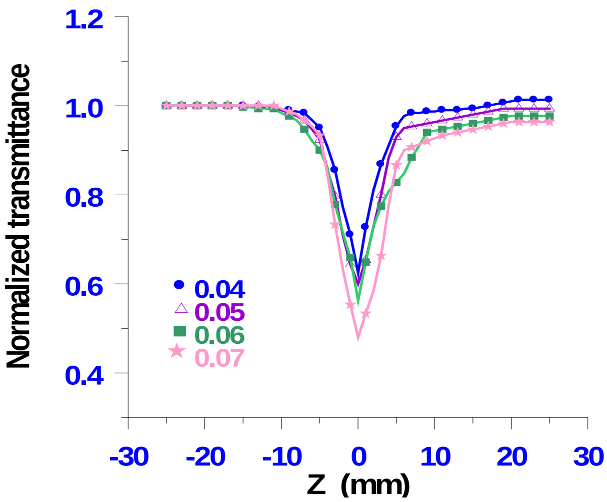


Figure 4 Open-aperture Z-scan data for the THF solution of the azo dye at different concentrations.

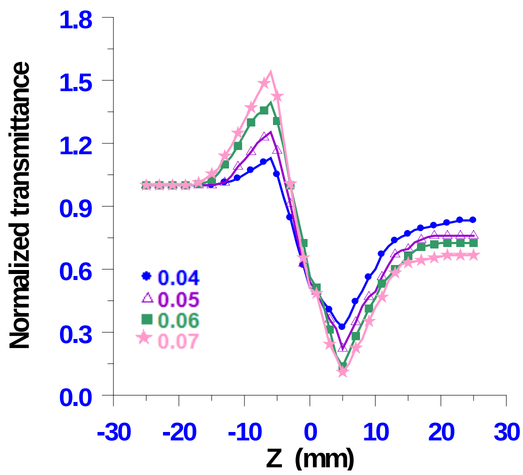


Figure 5 Closed-aperture Z-scan data for the THF solution of the azo dye at different concentrations.

The measurable quantity ΔT_{P-V} can be defined as the difference between the normalized peak and valley transmittances, $T_P - T_V$. The variation of this quantity as a function of $|\Delta\Phi_0|$ is given by [8]

$$\Delta T_{P-V} = 0.406(1-S)^{0.25}|\Delta\Phi_0| \quad (1)$$

Z-scan with a fully open aperture (S=1) is insensitive to nonlinear refraction (thin sample approximation). The aperture linear transmittance is given by $S = 1-\exp(-2r_a/\omega_a)$, with r_a =2.5 mm the aperture radius ,ω_a = 5 mm the radius of the laser spot before the aperture, $\Delta\Phi_0$ is the on-axis phase shift. The on axis phase shift is related to the third-order nonlinear refractive index by

$$\Delta\Phi_0 = kn_2 I_0 L_{eff} \qquad (2)$$

where $k = 2\pi/\lambda$ is the wave number and $L_{eff} = (1-\exp(-\alpha_0 L))/\alpha_0$ is the effective thickness of the sample , α is the linear absorption coefficient, L the thickness of the sample, I_0 the on-axis irradiance at focus and n_2 the third-order nonlinear refractive index. The defocusing effect of the dye in solution at various concentrations is shown in Figure 5. This defocusing effect is attributed to a thermal nonlinearity resulting from absorption of radiation at 635 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 severe phase distortion of the propagating beam.

Generally the measurements of the normalized transmittance versus sample position, for the cases of closed and open aperture, allow the determination of the nonlinear refractive index , n_2, and the reversible saturation absorption (RSA) nonlinear coefficient, β [9,10]. Here, since the closed aperture transmittance is affected by the nonlinear refraction and absorption, the determination of n_2 is less straightforward from the closed aperture scans. Therefore, it is necessary to separate the effect of nonlinear refraction from that of the nonlinear absorption. A simple and approximate method [11] to obtain purely effective n_2 is to divide the closed aperture transmittance by the corresponding open aperture scans (see Figure 6).

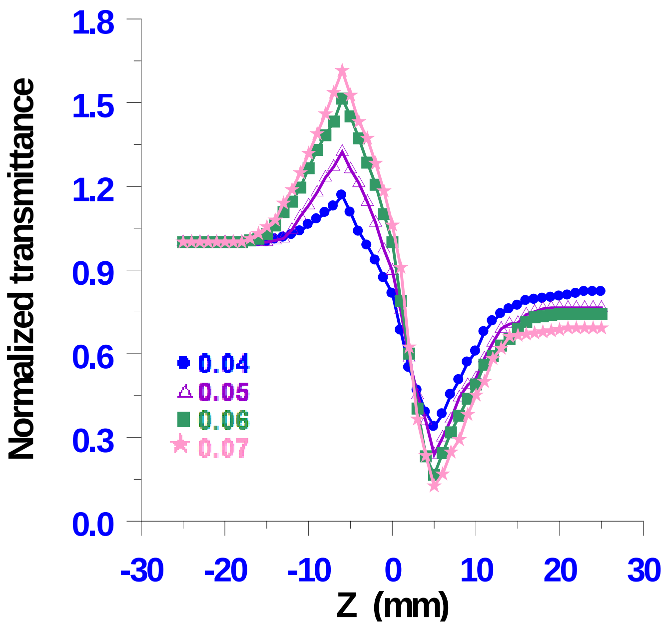


Figure 6 Pure nonlinear refraction curve of dye in solvent at various concentrations.

With an open aperture the sample's transmittance is related to the nonlinear absorption coefficient through the relation [12]:

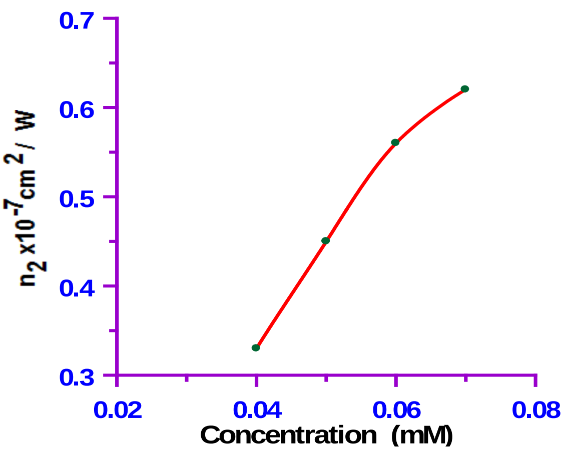
$$\beta = \frac{2\sqrt{2}\Delta T}{I_{\circ} L_{eff}} \qquad (3)$$

in which β is the nonlinear absorption coefficient, α is the linear absorption coefficient, L is the thickness of the sample, I_0 is the intensity of the laser beam at the focus (z = 0), and z_0 is the Rayleigh radius. In the Gaussian approximation, z_0 is related to the beam waist through the relation $z_0 = \pi\omega_0^2/\lambda$, where λ is the wavelength. The experiment was repeated for the pure solvent (Tetrahydrofuran) to account for its contribution, but no significant measurable signals were produced in either the open or the closed Z-scan traces. The nonlinear parameters calculated are as tabulated in Table 1.

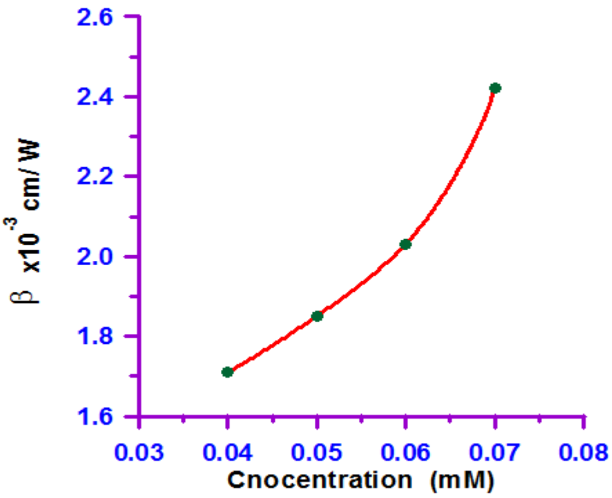
Table 1 Nonlinear parameters of janus green in tetrahydrofuran

Concentration(mM) (Liquid medium)	Δn $\times 10^{-4}$	$n_2\ cm^2/W$ $\times 10^{-7}$	$\beta\ \ cm/W$ $\times 10^{-3}$
0.04	2.29	0.33	1.71
0.05	3.09	0.45	1.85
0.06	3.85	0.56	2.03
0.07	4.26	0.62	2.42

From Figure 7a, b there is an increasing trend for the values of n_2 and β as the concentration increases. This may be attributed to the fact that, as the number of dye molecules increases when concentration increases, more particles are thermally agitated resulting in an enhanced effect. The optical nonlinearity of the dye 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.



(a)



(b)

Figure 7 Concentration dependence of (a): n_2, (b): β of janus green dye in tetrahydrofuran

Conclusion

we have measured the nonlinear refraction index n_2 and the nonlinear absorption coefficient β for the solutions of janus green for various concentrations using the Z-scan technique with 635 nm of cw laser. 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. All the solutions samples showed a large nonlinear refractive index of the order of 10^{-7} cm^2/W and 10^{-3} cm/W, respectively. All these experimental results show that the solution of janus green is a promising material for applications in nonlinear optical devices.

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دراسة الخواص البصريه اللاخطيه لمحاليل باستخدام تاثير ليزر مستمر

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قسم الفيزياء/ كلية التربيه/ جامعة البصره

الخلاصـه:

دراسة الخواص البصريه اللاخطية لمحاليل صبغة الجينيس الزرقاء(صبغه الازو) المذابة في رباعي هيدرو الفوران والتي تمت بواسطة تقنية المسح البصري باتجاه محور Z بأستخدام ليزر ذي النبضه المستمر بطول موجي قدره 635 نانومتر .وقد وجد ان معامل الانكسار اللاخطي ومعامل الامتصاص اللاخطي بحدود 10^{-7} سنتمتر مربع/ الواط و 10^{-3} سنتمتر/واط على التوالي. أظهرت هذه النتائج ان لهذه العينه تطبيقات في مجال البصريات اللاخطيه.وجميع العينات حضرت ودرست مباشره.