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Z-scan measurement for the nonlinear absorption and the nonlinear refraction of poly1,4-diazophenylene-bridged-tris(8-hydroxy-quinoline) aluminum ($PDPAlq_3$)

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ABSTRACT

The nonlinear optical properties of the poly1,4-diazophenylene-bridged-tris(8-hydroxy-quinoline) aluminum ($PDPAlq_3$) solution were studied using single beam Z-scan technique with a continuous-wave Diode laser radiation at 657.2 nm with 10 Hz repetition rate. The results show that the solution of $PDPAlq_3$ exhibits large nonlinear refractive index ($n_2 = -1.7642 \times 10^{-12}$ m^2/W) and nonlinear absorption coefficient ($\beta = 1.12 \times 10^{-6}$ m/W). The negative sign of the nonlinear refractive index n_2 indicates that the material exhibits self-defocusing optical nonlinearity. The evaluation of the figure of merit ($W = 1.8$) shows that the solution of $PDPAlq_3$ is sufficient for application in all-optical switching technology. These results show that the solution of $PDPAlq_3$ have potential application in nonlinear optics.

1. Introduction

In recent years, great effort has been made in order to explain the behavior of light beams propagating through interfaces separating optical media with different nonlinear refractive indexes. Development of high power laser sources has motivated an extensive research in the study of nonlinear optical properties and optical limiting behavior of materials [1].

Organic molecules with high nonlinear optical properties are required for photonic applications including optical switching, data storage and optical information processing. The study of linear and nonlinear optical coefficients is very important to tune the nonlinear optical (NLO) properties by the appropriate design of organic systems at the molecular level [2–5].

Nonlinear absorption can be classified into two types: first, transmittance increases with increasing optical intensity, this nonlinear absorption corresponds to saturable absorption (SA). Second transmittance reduces with increasing optical intensity; this nonlinear absorption includes two photon absorption (TPA) and reverse saturable absorption (RSA) [6,7].

The physical origin of nonlinear refraction can be electronic, molecular, electrostrictive or thermal. The first optical limiter was based on thermal mechanism with a CW laser; the thermal effects have been shown to be efficient even with nanosecond pulses [8–10].

Several techniques have been developed to measure the nonlinear optical refractive index n_2 (nonlinear interferometry, degenerate four-wave mixing and ellipse rotation, among other). These techniques are sensitive but usually require relative complex experimental [11–16]. One of the most important techniques to measure nonlinear refractive index was showed by Sheik Bahaei et al. this technique is simple and versatile yet is highly sensitive. Where one measures the change in transmittance of a focused laser beam through sample that is being moved through the focal point of the lens. Fig. 3 shows the set up of this technique [17]. An accurate knowledge of linear absorption coefficient (α_o) is necessary for the use of this technique, an easy way to measure α_o is to use the Bear's low [18].

In this work, we report the experimental measurements of the nonlinear refractive index n_2, nonlinear absorption coefficient β for poly1,4-diazophenylene-bridged-tris(8-hydroxy-quinoline) aluminum ($PDPAlq_3$) solution by using Z-scan technique with a continuous-wave Diode laser radiation at 657.2 nm with 10 Hz repetition rate.

2. Synthesis of poly1,4-diazophenylene-bridged-tris(8-hydroxy-quinoline) aluminum ($PDPAlq_3$)

The synthesis of polymer is as stated in Ref. [19], 0.63 mml of (1,4-diamino benzene)($C_6H_8N_2$) was mixed in 2.5 ml of water and 20 ml concentration hydrochloric acid (HCl) with heating. The mixture was cooled in iced bath for 10 min. 0.7 g sodium nitrite ($NaNO_2$) was solvated in 5 ml water and cooled in iced bath for 5 min. A

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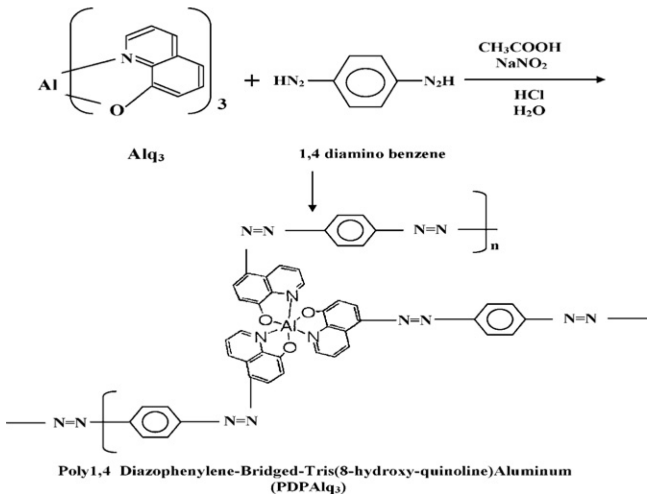

Fig. 1. The reaction equation of poly1,4-diazophenylene-bridged-tris(8-hydroxy-quinoline) aluminum ($PDPAlq_3$).

0.5 g Alq3 and 4 g sodium acetate ($CH_3COO^-Na^+$) in 50 ml water was added to the $NaNO_2$ solution and the mixture was shacked and added to the amino solution in ice bath (273–278 K) and then heated for 30 min at 358 K, the reaction takes place as shown in Fig. 1. The result was analyzed and characterized.

The spectral properties of the $PDPAlq_3$ solution is studied by recording the absorption spectra of the solution using an Aquarius double beam UV: Spectrophotometers (CE-7200), the absorption spectrum of the aqueous solution is shown in Fig. 2.

3. Nonlinear studies

The nonlinear response of $PDPAlq_3$ was characterized using the Z-scan technique. This technique employs a single Gaussian laser beam in a tight focus geometry to measure the transmittance of a nonlinear medium as a function of the sample position [20]. Z-scan technique is simple and effective tool for determining the nonlinear properties of various kinds of materials, because it provides the magnitude and the sign of the nonlinear index of refraction n_2, and also the nonlinear absorption coefficient β. In this technique, a polarized Gaussian laser beam, propagation in the Z-direction, is focused to a narrow waist. The sample is moved along the Z-direction and the transmitted intensity is measured through a finite aperture in the far field as a function of the sample position Z, measured with respect to the focal plane. As the sample moves through the beam focus (at $Z = 0$), self-focusing or self-defocusing modifies the wave front phase, thereby modifying the detected beam intensity [17,20]. The basic Z-scan setup is shown in Fig. 3.

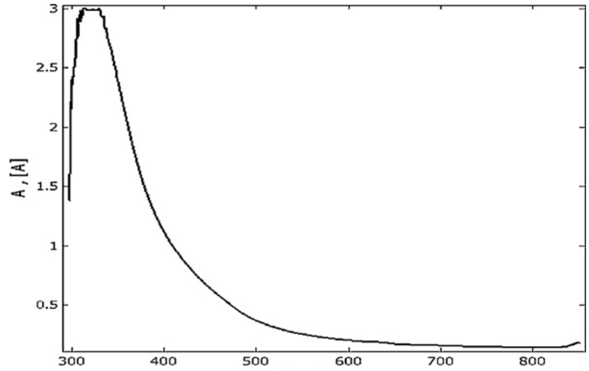

Fig. 2. The absorption spectrum of $PDPAlq_3$ solution.

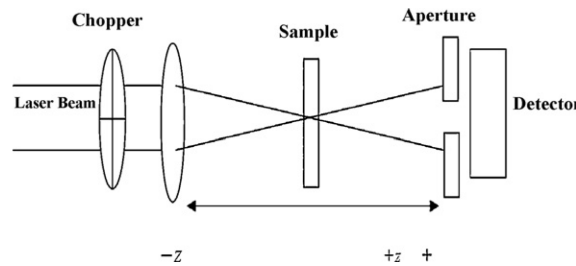

Fig. 3. The basic Z-scan setup.

The Z-scan experiments were performed using a Diode laser beam (657.2 nm with 10 Hz repetition rate, power 25 mW) as the light source, which was focused by +50 mm focal length lens. The laser beam waist ω_o at the focus is measured to be 19.39 μm and the Rayleigh length z_o = 1.79 mm. The schematic of the experimental set up used is shown in Fig. 3. A 1 mm wide optical cell containing the $PDPAlq_3$ solution is translated across a focal region along the axial direction of the propagation laser beam. The transmission of the beam through an aperture placed in the far field was measured using photodetector fed to the power meter.

4. Results and discussion

When the Gaussian beam illuminates the medium, the medium absorbs the light and its temperature rises, the raise in temperature results in change of refractive index and induces self-diffraction [21]. The formation of the spatial ring pattern is attributed to induced spatial self-phase modulation arising from laser induced refractive index change and thermal lensing [22]. The spot of the transmitted beam was photographed at far away distance from the sample, when the sample was at different positions. Fig. 4(a)–(d) shows the transmitted beam profiles and Fig. 5(a)–(d) shows the distribution of intensity for $PDPAlq_3$ solution, when the sample was far from the focus ($Z = -10$ mm), at the focus ($Z = -4$ mm), out of focus ($Z = 0$ mm) and away from the focus ($Z = +10$ mm). Figs.4(b) and 5(b) show that the spot of the transmitted beam has minimum size only when the sample was at the focus, Figs. 4(c) and 5(c) show the spatial ring pattern(self-diffraction).

The third order nonlinear refractive index n_2 and nonlinear absorption coefficient β of the $PDPAlq_3$ solution were determined from the open and close aperture Z-scan measurements, respectively. Fig. 6(a) and (b) shows the open aperture (OA) and closed aperture (CA) Z-scan data for $PDPAlq_3$ solution at incident intensity I_o = 4.24 kWatt/cm^2. A signature of peak-valley indicating a negative type of nonlinearity from closed aperture (self-defocusing occurs) and two photon absorption type behavior for open aperture scan was absorbed.

The change in the refractive index n_2 can be evaluated by use of the difference in the peak-valley normalized transmittance $\Delta T = T_p - T_v$, which is given by [20]:

$$\Delta T = 0.406(1 - S)^{0.25}|\Delta\varphi_o| \qquad (1)$$

where $\Delta\varphi_o$ is the on-axis phase shift. The on-axis phase shift is related to the third order non-linear refractive index by [20]:

$$\Delta\varphi_o = kn_2 I_o L_{eff} \qquad (2)$$

where $S = 1 - \exp(-2r_a/\omega_a)$ is the aperture linear transmittance ($S = 0.39$) with r_a denoting the aperture radios and ω_a denoting beam radius at the aperture in the linear region, I_o is the intensity of the laser beam at focus $Z = 0$, $L_{eff} = (1 - \exp(-\alpha_o L))/\alpha_o$ is the effective thickness of the sample (L is sample thickness), (α_o) is linear absorption coefficient of the sample and $k = 2\pi/\lambda$ is wave number.

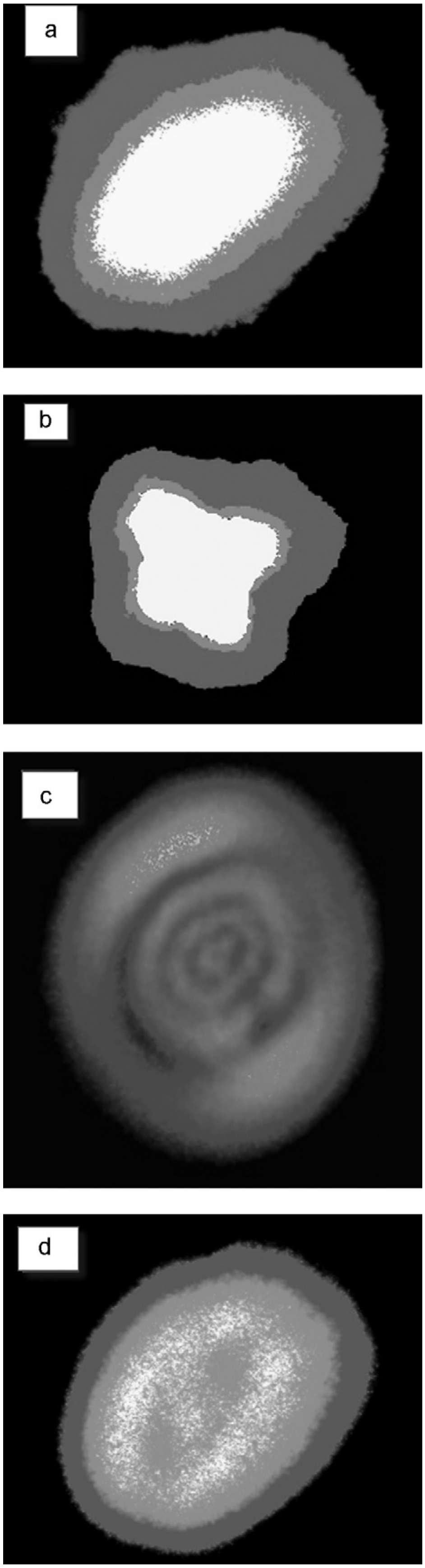

Fig. 4. The transmitted beam profiles corresponding to the sample positions: (a) far from the focus, (b) at the focus, (c) out of focus and (d) away from the focus for $PDPAlq_3$ solution.

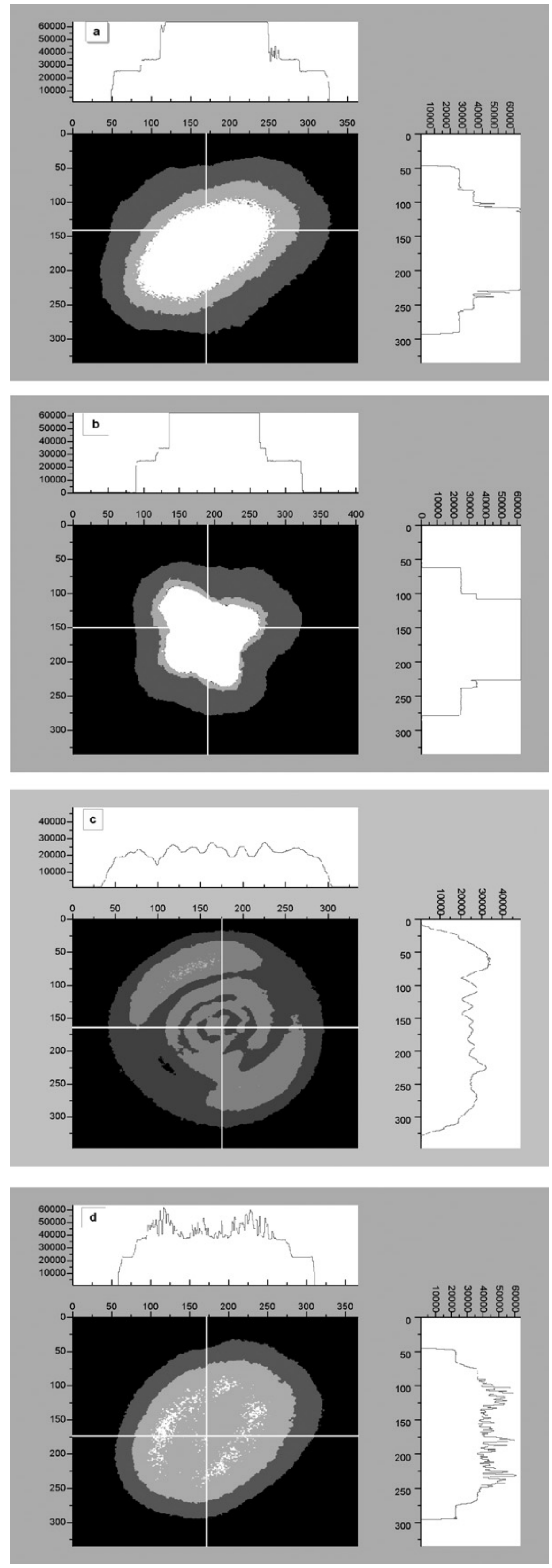

Fig. 5. The transmitted beam profiles and the distribution of intensity corresponding to the sample positions: (a) far from the focus, (b) at the focus, (c) out of focus and (d) away from the focus for $PDPAlq_3$ solution.

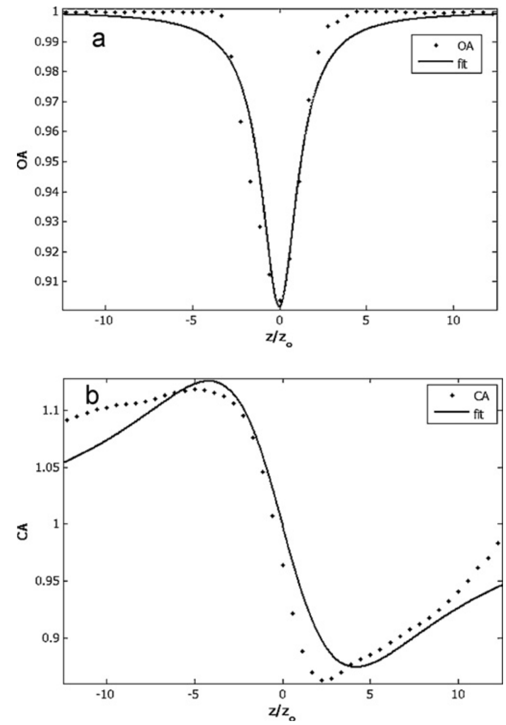

Fig. 6. Represent Z-scan experimental data (square) for PDPAlq$_3$ solution: (a) open aperture and (b) close aperture, solid line shows theoretical fit to the experimental data.

The normalized transmittance of pure nonlinear refraction is given by [17]:

$$T(z) = 1 + \frac{4x\Delta\varphi_o}{(x^2+9)(x^2+1)} \quad (3)$$

where $x = z/z_o$, $z_o = \pi\omega_0^2/\lambda$ is the diffraction length of the laser beam, and ω_o is laser beam waist at the focal point.

From Eqs. (1) and (2), the nonlinear refractive index n_2 can be obtained and the corresponding changes in the refractive index $\Delta n = n_2 I_o$. The theoretical fit results showed that the peak to valley ΔT_{P-V} indicates the negative sign of nonlinear refractive index, n_2 (self-defocusing) for PDPAlq$_3$ solution. Closed aperture data shows that the peak-valley normalized transmittance $\Delta T = 0.255$.

The nonlinear absorption coefficient β can be estimated from the open aperture Z-scan data using the relation [20]:

$$T(z,\ s=1) = \sum_{m=0}^{\infty} \frac{[-q_o(z)]^m}{(m+1)^{3/2}} \quad \text{For } |q_o(0)| < 1 \quad (4)$$

where $q_o(z) = \beta I_o L_{eff}/(1+z^2/z_o^2)$, $z_o = \pi\omega_0^2/\lambda$ is the diffraction length of the laser beam, and ω_o is laser beam waist at the focal point. The measurement details and the results are listed in Table 1.

For practical use in ultra-fast all-optical switching devices many consideration have been taken into to investigate the effectiveness of nonlinear materials. The figure of merit W has to be satisfied for 2π phase shift in order to evaluate its application in such devices [23]:

$$W = \frac{\Delta n_{\max}}{\alpha\lambda} > 1\,, \quad T = \frac{2\beta\lambda}{|n_2|} < 1 \quad (5)$$

Under 657.2 nm excitation, for PDPAlq$_3$ solution, $W = 1.8$ and $T = 0.83$, which indicates that the nonlinear optical properties of PDPAlq$_3$ solution are sufficient for application in all-optical switching technology. The fast response time and large nonlinear refraction makes PDPAlq$_3$ solution promising for use in non linear optical devices.

Table 1
The measurement details and the results of the Z-scan technique.

Laser beam wavelength (λ)	657.2 nm
Lens focal length (f)	+50 mm
Optical path distance (Z)	15 cm
Spot-size diameter in front of the aperture (ω_a)	1.2 cm
Aperture radius (r_a)	2.5 mm
Incident intensity at the focus ($Z=0$)	4.24 kW/cm^2
Effective thickness (L_{eff})	0.994 mm
Linear absorption coefficient (α_o)	0.105 (1/cm)
Nonlinear refractive index (n_2)	-1.7642×10^{-12} m^2/W
Nonlinear absorption coefficient (β)	1.12×10^{-6} m/W
The change in the refractive index (Δn)	-7.4834×10^{-5}

5. Conclusion

The third-order nonlinear refractive index n_2, and nonlinear absorption coefficient β, of poly1,4-diazophenylene-bridged-tris(8-hydroxy-quinoline) aluminum (PDPAlq$_3$) solution was studied using a single beam Z-scan technique under continuous-wave Diode laser radiation at 657.2 nm with 10 Hz repetition rate. The Z-scan studied showed that the PDPAlq$_3$ solution exhibited negative sign of nonlinear refractive index (self-defocusing) and nonlinear absorption includes two photon absorption (TPA), for the non-resonant absorption. The n_2, β and Δn values were found to be -1.7642×10^{-12} m^2/W, 1.12×10^{-6} m/W and -7.4834×10^{-5}. The evaluation of the figure of merit W shows that the solution of PDPAlq$_3$ is sufficient for application in all-optical switching technology.

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