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The Effects of Initial Laser Intensity on the Nonlinear Optical Properties of The Laser Dye DQOCI Doped Films Using Z-Scan Technique

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Abstract:

This study is dedicated to investigate the effects of initial laser intensity on the nonlinear optical properties of the laser dye DQOCI dissolved in methanol with a concentration of 10^{-5} M and doped with PMMA film. The properties were studied by using open and closed aperture Z-scan technique, with different levels of initial intensity (I_0), excited by continuous diode solid-state laser at a wavelength of 532 nm. Three lenses of different focal lengths were employed to change the radius of the Gaussian laser beam and then change the initial intensity. For I_0= 6.83 and 27.304 kWatt/cm^2, the Z-scan curves show a saturation of absorption (SA) known as the negative type of nonlinearity, in which the absorption coefficient β_2 decreases and the transmittance increases with increasing the initial laser intensity. With I_0 equal to 3.03 KWatt/cm^2, the nonlinear absorption changes from SA to RSA, where the transmittances is reduced with the increase of intensity (z□0) as analyzed by the theory of free carrier nonlinearities. The closed aperture z-scan shows a pre-focal transmittance minimum (valley) and a post focal transmittance maximum (peak) which reflects the z-scan signature of a positive nonlinearity (self-focusing) due to Kerr effect. Each of nonlinear refractive index (n_2), nonlinear absorption coefficient (β_2), and third-order nonlinear optical susceptibility (χ^3) are intensity-dependent.

Keywords: Nonlinear Optical Properties, Dye Doped Polymer Films, Z-Scan Technique**.**

تأثير شدة الليزر الاولية على الصفات البصرية اللاخطية لأغشية الصبغة الليزريةDQOCIباستخدام تقنية المسح على المحور الثالث

امل فيصل جعفر

المعهد التقني الطبي/المنصور, الجامعه التقنية الطبية, بغداد, العراق

الخلاصة:

هذه الدراسة حول تأثير اشعة الليزر الاولية على الصفات البصرية اللاخطية لأغشية الصبغة الليزرية DQOCI المذابة في الميثانول وبتركيز 10^{-5}مولاري والمطعمة مع PMMA باستخدام تقنية المسح على المحور الثالث باستخدام فتحة وبدون استخدام الفتحة وباستخدام شدات ليزرية اولية مختلفة ومصدر الاثارة هو ليزردايود الحالة الصلبة المستمر ذو الطول الموجي 532 نانومتر وباستخدام ثلاث عدسات ذات بعد بؤري مختلف لأجل تغيير نصف قطر حزمة الليزر الكاوسية وبذلك تتغير الشدة الاولية. للشدة الاولية I_0= 6.83 كيلو وات/سم^2 و 27.304 كيلو وات/سم^2 أظهر المسح على المحور الثالث تشبع في الامتصاص يعرف بالنوع اللاخطي السالب حيث يقل معامل الامتصاص اللاخطي (β_2) وتزداد النفاذية بزيادة شدة الليزر

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الاولية. مع الشدة الاولية 3.303 كيلو واط/سم^2 يتغير الامتصاص اللاخطي من الامتصاص المشبع الى الامتصاص المشبع المعكوس حيث تقل النفاذية بزيادة الشدة الاولية عند z=0 والتي فسرت اعتمادا على نظرية الحاملات الحرة اللاخطية . في حالة استخدام الفتحة اظهر المسح على المحور الثالث انخفاض بالنفاذية قبل البؤرة (وادي) وبعد البؤرة تكون النفاذية اعلى مايكون (قمة)وهو دليل على ان المسح على المحور الثالث هو من النوع الموجب (تركيز –ذاتي) نتيجة لظاهرة كير. كل من معامل الامتصاص اللاخطي (β_2) ,معامل الانكسار (n_2)اللاخطي و الحساسية البصرية اللاخطية (χ^3) مرتبطة مع الشدة الاولية.

1-Introdaction

The laser dye 1,3'-Diethyl-4,2'-quinolyloxacarbocyanine Iodide (DQOCI) is a large organic molecule with a molecular weight of 470.35 m_u. [1, 2]. When this laser dye was doped with a polymer like PMMA, it provided a tunable range of laser wavelengths that covered electromagnetic spectra starting from UV through visible to infrared with high quantum efficiencies [3]. It caused important changes in their physical properties [4] and exhibited exceptional nonlinear optical properties. Dye doped polymer material matrix can increase the absorptivity or fluorescence as well as the opto-chemical and opto-physical stability [5].

There are many different techniques which are used to prepare polymer films [6], one of them being the spin coating technique. Spin coating is the predominating technique used to produce uniform thin films from the photosensitive organic materials with thickness of the order of micrometers and nanometers. This process has been widely used in the fabrication of optical mirrors, integrated circuits, magnetic disks for data storage, and color television screens [7].

The discovery of third order nonlinear optical properties (NLO) of materials, such as self-focusing, self-defocusing, two-photon absorption, reverse saturated absorption, and nonlinear scattering, assisted the development and expansion of its applications, including optical limiting devices, Q-switch, passive mode locking, optical operation, and light storage, etc. [5].

The nonlinear optical properties of DQOCI films doped with the polymer PMMA were studied by using z-scan technique which is a single-beam technique that aids to determine both the sign and magnitude of refractive index, nonlinear absorption coefficient and third order nonlinear susceptibility. This method is simple, rapid, and accurate [8, 9]. Z-scan technique depends on the usage of Gaussian laser beam. There are two parts of the process of measuring the normalized transmittance as a function of *Z* (Z scan). The first is the open aperture *(*no aperture) Z scan, which is sensitive only to the induced changes in absorption, whereas the second is the closed-aperture Z scan, which displays the induced refractive changes.

“For a material that has a strong nonlinear effect, as the light intensity is increased the absorption of light increases too, and beyond a certain input intensity the output intensity approaches a constant value. These materials can be used for limiting the amount of optical power entering a system. [10]. For estimating the advantage of an optical limiting material, it should exhibit broad band spectral responses, i.e., it is transparent at low intensities while exhibiting a large nonlinearity at high intensities over a broad band spectral range [11].

In this paper, the optical nonlinearity (nonlinear refraction ,nonlinear absorption and third order susceptibility) of DQOCI in methanol solvent and a thin film of PMMA was studied at 50 mW solid state laser power at a wavelength of 532 nm.

2. PRACTICAL PART

2.1. Materials preparation

In this work, the organic dye, laser dyeDQOCI ($C_{23}H_{23}N_2OI$) from Lambdachrome was used, and its molecular structure is shown in Figure-1.

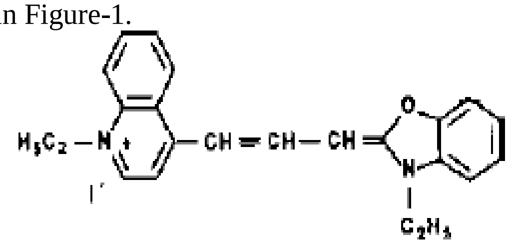


Figure 1- The molecular structure of the organic dye DQOCI [1]

Dye solutions of a concentration of 10^{-5} M in Methanol (Lab-Scan LTd., Analitical Science HPLC Ireland-Dublin) was prepared by weighting an amount of the dye with a matter balance of a sensitivity of 10^{-4} gm and doping it dye with the polymer PMMA from ICI Company. Polymer solution was prepared by dissolving the required amount of polymer (7 gm) in 100 ml (7%w/v) of methanol.

The doped films (with a ratio 25% of 2 ml of DODQI solution to 8 ml of the polymer solution) were prepared by the spin coating method using a Spin Coater from Holmarc Opto-Mechatronics PVT-Ltd, India, at 2000 r/m for 30 sec. The film was dried for 24 hr at room temperature. The thickness of the films was measured with (Mini-test 3000 microprocessor (Electrophyisk, Germany, ERICHSEN) yielding a film of 21.5 µm thicknesses.

2.2. UV-Vis Spectroscopic Characterization

The absorption spectra of DQOCI film doped with PMMA were recorded by a visible Shimadzu spectrophotometer. (UV 160), which operates in a wavelength range of 200 to 1100 nm at a scanning speed of 1500 nm/min.

Figure-2 shows the absorption spectra of DQOCI solution of the concentration of 10^{-5} M and the film doped with PMMA. It is clear from the graphs that the peak of the spectrum (λ_{max}) of the doped film had a red shift (toward the long wavelengths) compared with the spectrum of DQOCI solution. The doping implies that an increased number of molecules per volume, conjugated via the pi-electron system, shifts the absorption maximum about 45 nm in the same direction, and moves the absorption maxima to longer wavelengths [12]. When the chromophores absorbed light over the range of wavelengths, they produced a range of energy jumps so that the rotations and vibrations energy of the molecules was changed, producing the gap between the two spectra and causing the red shift. The presence of DQOCI enhances the UV absorption in the doping films and modifies the optical behavior of the doped polymer film. Each additional double bond in the conjugated π-electron system shifts the absorption maximum λ_{max} in the same direction and increases the molar absorptivity of each newly conjugated double bond [13].

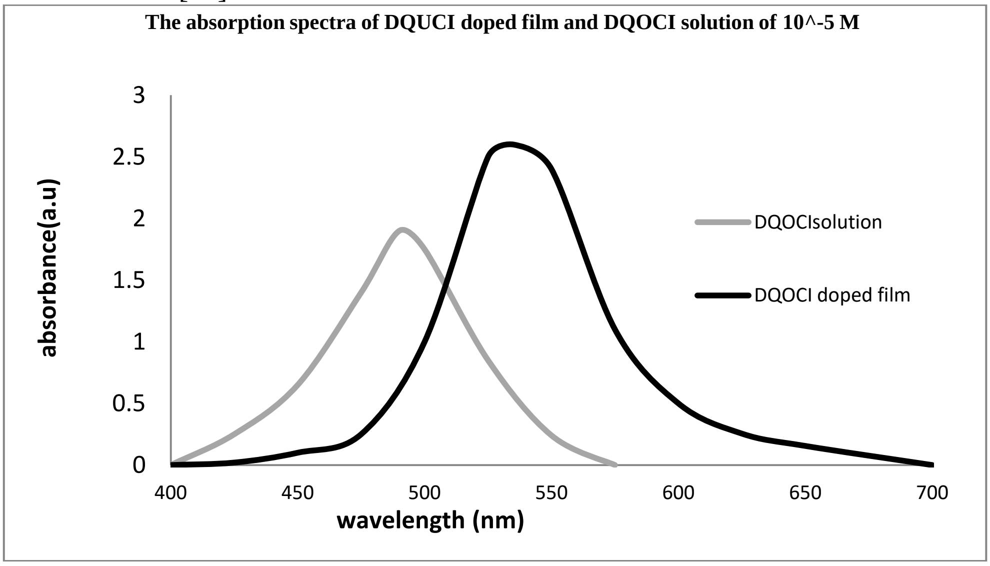
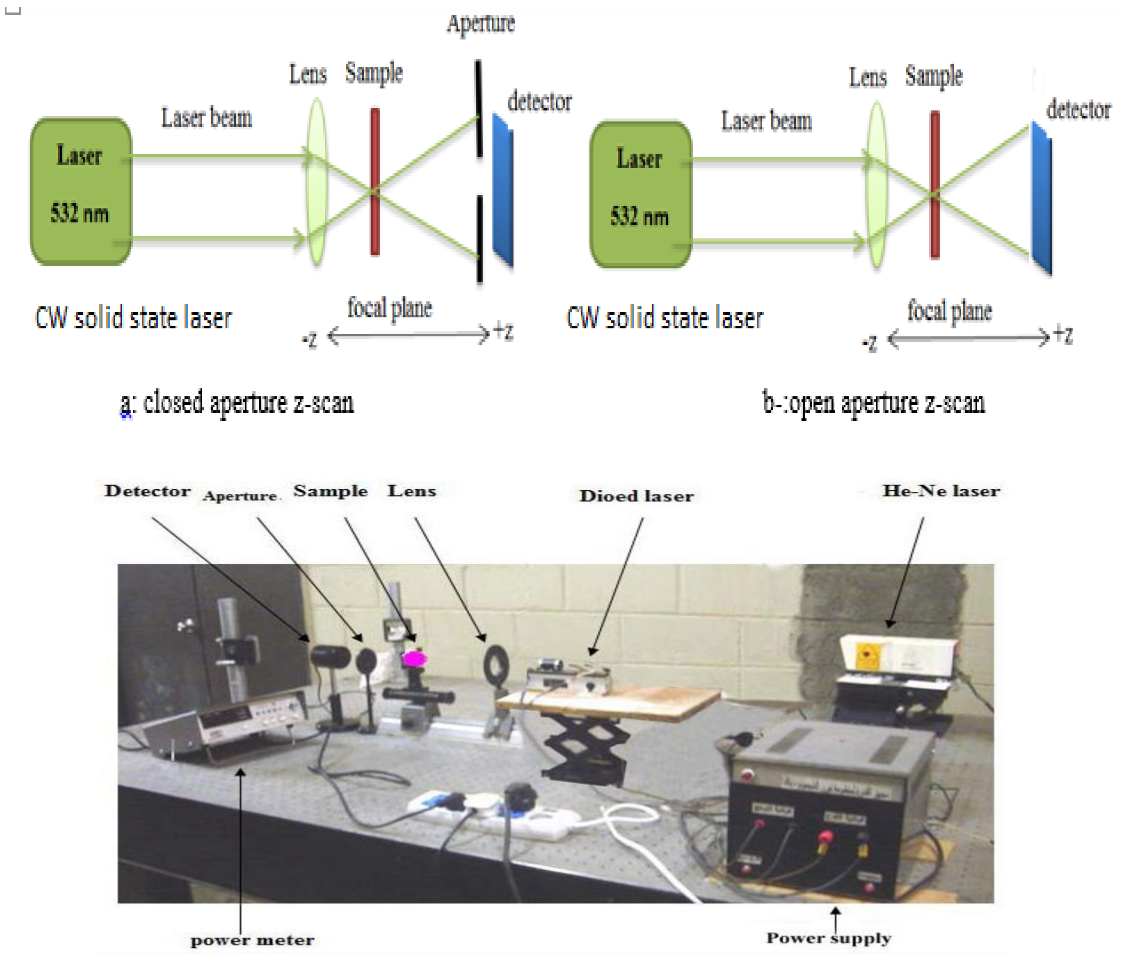


Figure 2- The absorption spectra of DQOCI doped film and DQOCI solution

2.3. Z-scan Experiment

Z- Scan technique was used to study the nonlinear interaction and response between DQOCI – PMMA doped films and the incident laser beam. A continuous solid-state laser at a wavelength of 532 nm was used as an excitation source. A Gaussian laser beam was obtained by using a converging lens with focal lengths (f) of 5, 10, and 15 cm, which accordingly resulted in Rayleigh length (z_0) values of 0.688, 2.753, and 6.196 mm, respectively, producing I_o (laser incident intensity) values of 27.304, 6.83, and 3.03 Kw/cm^2, respectively.

Figure-3 Shows the setup of the Z-Scan experiment.



c: Photographic illustration for the experiment setup
Figure 3-Z-scan experiment setup [10].

The experiments were conducted by moving the sample (DQOCI doped films) in the focal plane and the focal point along the propagation of the laser beam (z-axis direction) in two experiments. The first was named as the closed aperture z-scan, for which the transmission of the laser beam was measured as a function of the sample position through -z to +z by the photo detector, as shown in fig. (3) a. Also, the nonlinear properties, namely the third order refractive index (n_2) and the real part of the third order susceptibility ($R\chi^{(3)}$) of the doped films were investigated. The second experiment is the open aperture z-scan, illustrated in Figure-(3 b), where the aperture in front of the detector was removed and a converging lens was used to collect the incident laser beam transmitted from the sample. With the open aperture z-scan, the nonlinear absorption coefficient (β_2) and imaginary parts of the third-order nonlinear optical susceptibility ($Im\chi^{(3)}$) of the doped films were measured. [14, 15]

The principle of the closed aperture Z-scan is based on the transformation of phase distortions to amplitude distortions when the sample moves through the beam propagation. The scanning starts from a distance far away from the focus (- z), while the beam irradiance is low, leading to linear transmittance and negligible nonlinear refraction. As the sample is brought closer to the focus, the beam irradiance increases, producing self-lensing in the sample. Self-lensing is a nonlinear optical phenomenon induced in the materials when it is exposed to an intense electromagnetic radiation, where the medium refractive index changes with the electric field intensity and, hence, it acts as a focusing lens [16, 17] .A negative self-lensing (self-defocusing) before the focus reduces the diffraction, leading to a small beam at the aperture and a raise of the transmittance. When the sample crosses the focal plane to the right (+ z), the same self-defocusing effect will tend to raise the diffraction and minimize the aperture transmittance, as demonstrated in Figure-4, A prefocal transmittance maximum (peak) and a post focal transmittance minimum (valley) will represent the z-scan signature of a negative nonlinearity, as shown by the solid line in Figure-4. While a positive one, following the same analogy, will give rise to an opposite valley-peak configuration, as shown by the dotted line in Figure-4 [18, 19].

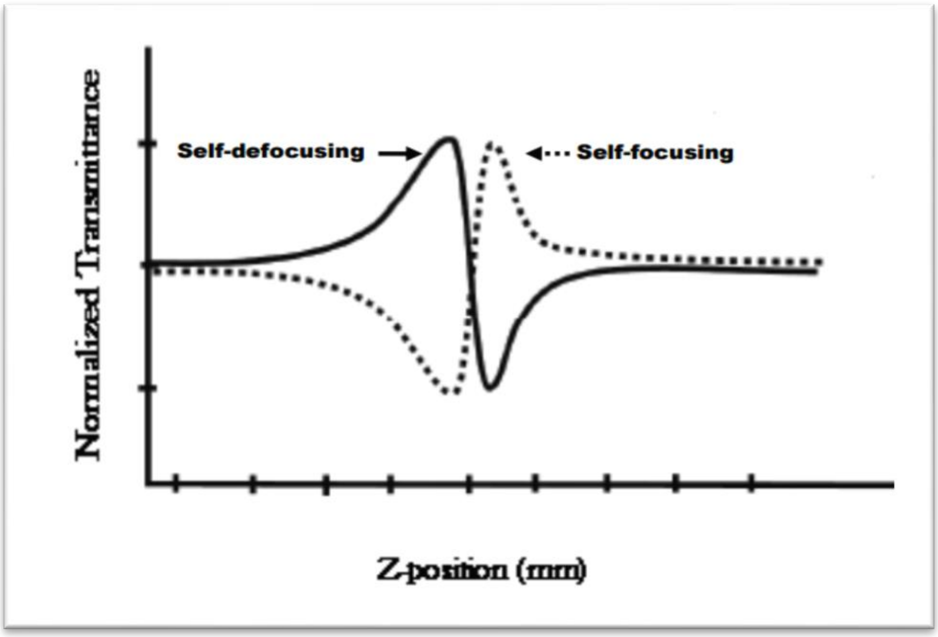


Figure 4-Theoretical Z-scan transmittance curves for a third order nonlinearity [19].

An open-aperture *Z*-Scan measures the change in intensity of a beam, focused by lens **l** in Figure-3, in the far field at the detector PD, which captures the entire beam and gives an estimate of the absorptive nonlinearity of a sample [20]. The change in intensity causes two photon absorption (TPA), multi-photon absorption, or saturation absorption (SA) in the sample as it travels through the beam waist. In the focal plane where the intensity is the greatest, the largest nonlinear absorption is observed. At the "tails" of the *Z*-scan signature, where |Z| >> Zo, the beam intensity is too weak to elicit nonlinear effects. The higher order of multi-photon absorption present in the measurement depends on the wavelength of light and the energy levels of the sample [19]. With SA, the absorption coefficient decreases resulting in the transmittance's increase with the increase in the input laser intensity. With reverse saturable absorption (RSA) or two photon absorption, the absorption coefficient increases resulting in the transmittance's decrease with the increase in the input laser intensity [10, 16].

3. Results and discussion

Figure-5 shows the results of the open aperture z-scan experiment for the laser dye DQOCI dissolved in methanol (10^{-5} M) with different I_0 values, doped with PMMA, and excited by continuous solid-state laser at a wavelength of 532 nm. Three lenses of different focal lengths were used to change the radius of the Gaussian laser beam and then change the initial intensity. For I_0=3.03 kWatt/cm^2 (f=15 cm) and I_0= 6.83 KWatt/cm^2 (f=10 cm), Z-scan curves show an upward peak, indicating a saturation of absorption (SA) that is known as the negative type of nonlinearity where the transmittance increased as the intensity increased (i.e. the absorbance decreased) [20]. When I_0 is 27.304 KWatt/cm^2 with a lens of a focal length of f=5 cm, the nonlinear absorption changes from SA to RSA or TPA, where the transmittances is reduced with the increase of intensity and the decrease of the focal length (Figure-6). All corresponding profiles are symmetric with respect to the focus (z = 0). Our results have a good agreement with those of an earlier report [21].

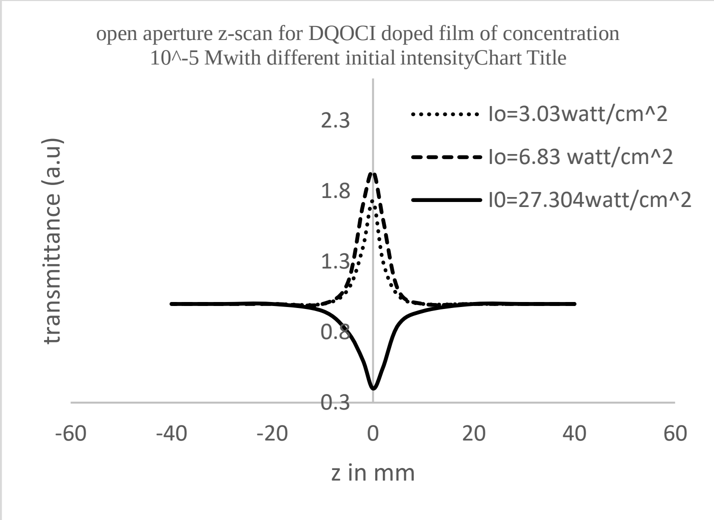


Figure 5-The open aperture z-scan experiment length and with different initial intensity

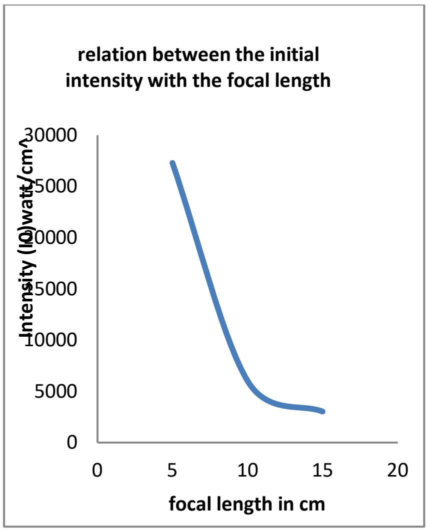


Figure 6-The relation between the focal the initial intensity

The measurements of the normalized transmittance against sample position z, for the case of open aperture, allowed determining the saturation absorption coefficient β. The nonlinear absorption coefficient β can be calculated from the open aperture Z-scan data by using equation (1):

$$\beta = 2\frac{2\sqrt{2\Delta T}}{I_0 L_{eff}} \qquad \ldots\ldots\ldots\ldots(1)\ [22]$$

where ΔT is the normalized transmittance, the value of which was taken from the open aperture saturation.
Figure-5.
L_{eff}. is the effective length of the sample, which can be determined from the following equation:

$$L_{eff} = [1-\exp(-\alpha_0 L)]/\alpha_0 \qquad \ldots\ldots\ldots\ldots (2), [23].$$

In this experiment. its value was 0.0022 cm.
α_0 is the linear absorption coefficient, which was found to be 28.87 cm^{-1}.

$$\alpha_0 = \frac{1}{L\ln(\frac{1}{T})} \qquad \ldots\ldots\ldots\ldots(3)\ , [24]$$

where L: sample thickness, T: linear transmittance, which was 0.98%. The initial intensity I_0 was measured by the equation:

$$I_0 = \frac{2p}{A} \qquad \ldots\ldots\ldots\ldots(4)$$

where p is the solid state laser power, which was 100 mWatt, whereas A is the laser spot area which can be calculated from the equation:

$$A = (\omega_o)^2\pi, \qquad \ldots\ldots\ldots\ldots(5)$$

where ω_o: the beam radius at the focal point,

$$\omega_o = \theta f \qquad \ldots\ldots\ldots(6)$$

where θ is the laser divergence and f is the focal length of the used lenses [24,25].
From equ.(6), it is clear that as (f) was increased, I_0 was decreased, as shown in Figure-6.
Calculating β_2 can help in determining the imaginary part of the third-order nonlinear optical susceptibility, i.e., $Im(\chi^3)$, as follows:

$$Im(\chi^3)\ (esu) = (10^{-2}\ c^2\ \varepsilon_0\ n_0\ \lambda^2\ k/4\pi)\ \beta_2(cm/W) \qquad \ldots\ldots\ldots(7)$$

with c, ε_0, n_0 ,λ and k refer to the light velocity in vacuum, electric permittivity, linear refractive index, and laser wavelength, respectively [26].
Table-1 shows the open aperture parameters of DQOCI doped films.

Table 1-Nonlinear open aperture parameters for PMMA film doped with DQOCI at a concentration of 10^{-5} M by using solid state laser at 530 nm with different initial intensity

f(cm) focal length	I_0 watt/cm^2	T(z)	β(cm/w)	Im χ^3 (esu)	Kind of absorption
15	3030	1.94	0.589	3.77	SA
10	6030	1.74	0.313	2.89	SA
5	27304	0.4	0.031	0.91	TPA

Table-1 shows the parameters of nonlinear absorption z-scan. For the doped film, it is clear that in case of saturation absorption, as I_o on axis irradiance increased, T(z) decreased, which led to decreasing the nonlinear absorption coefficient (β_2). The result in the case of SA is attributed to the fact that the sample bandgap was small and the was SA induced by one photon absorption which took place when the transmission was enhanced at focus (z = 0), in which β_2 decreased and the transmittance increased where the molecules were excited from the ground state to a higher state. Most of the molecules occupied the excited state. As the ground state is bleached, the system becomes increasingly transparent to the incident laser at 532 nm, resulting in a saturation of absorption [20]. This implies that the electrons on the valence band are transited to the conduction band and the laser intensity depletes many electrons from the valence band to the conduction band, leading to decreased absorbance [21]. This kind of action has useful applications in optical switching [22] and is well suited for passive Q-switching or mode locking of lasers [23].

The transformation from SA to RSA suggests that another nonlinear process takes place and becomes dominant, which could be probably due to the TPA (positive absorption). The minimum transmittance highlights the better optical limiting efficiency and can be used in efficient optical limiters [22] which can be exploited against radiation induced damage for the protection of eyes, light-sensitive sensors and CCD cameras.

Figure-7 shows the relation between I_0 and the nonlinear absorption coefficient and indicates an inverse relation with a statistical correlation factor equal to -0.919.

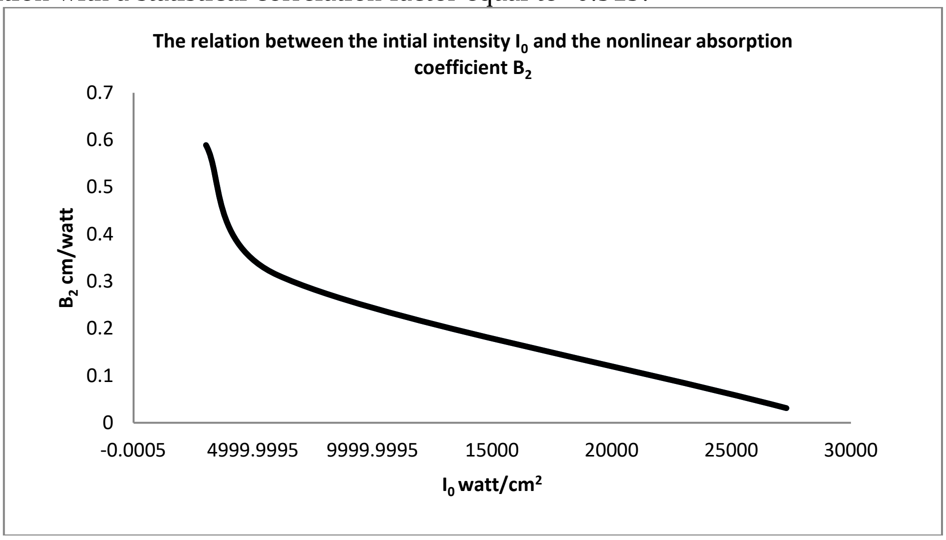


Figure 7-The relation between I_0 and the nonlinear absorption coefficient

Figure-8 shows the closed aperture z-scan experiment for the laser dye DQOCI dissolved in methanol with a concentration of 10^{-5}M, doped with PMMA and excited by continuous solid-state laser at a wavelength of 532 nm with different initial intensity values at the focus.

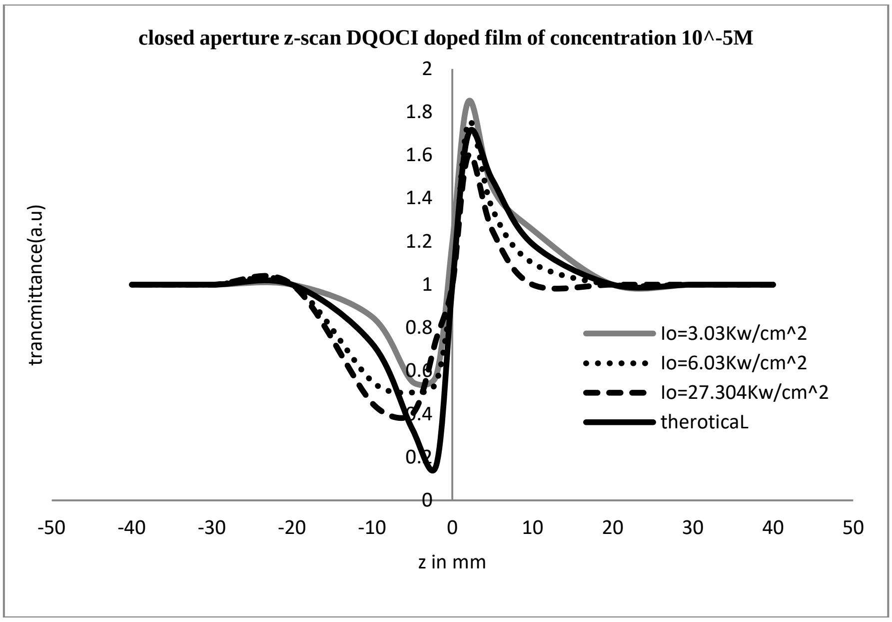


Figure 8-The closed aperture z-scan experiment for the laser dye DQOCI dissolved in methanol (10^{-5}M), doped with the polymer PMMA, and excited by continuous solid-state laser at a wavelength of 532 nm with different initial intensities at the focus.

The nonlinear refraction can be extracted from the division of the closed aperture reading by the open aperture reading [27, 28], Figure-8 shows a pre-focal transmittance minimum (valley) and a post focal transmittance maximum (peak), which reflects the z-scan signature of a positive nonlinearity (self-focusing) that is fitting with the dotted line in Figure-4 and solid black line in Figure-8. This behavior is attributed to the variation of refractive index with temperature, resulting in the typical shape of a *Z*-scan trace for Kerr nonlinearity. The energy from the focused laser beam is transferred to the sample through linear absorption and is manifested in terms of heating the medium, leading to a temperature gradient. Thereby the refractive index changes across the sample which then acts as a lens (Kerr effect) [29-31].

Table 2-shows the results of the nonlinear parameters of the closed aperture z-scan for the doped DQOCI

I_0 W/cm^2 DQOCI	$T_{max.}$	$T_{min.}$	ΔT	$\Delta\phi_o$	n_2	RX^3(esu)	ImX_3	X_3
3030	1.6	0.4	1.2	2.96	3.964E-06	2.05E-02	0.255	2.56E-01
6030	1.75	0.5	1.25	3.079	2.075E-06	1.07E-02	0.10991	1.10E-01
27304	1.85	0.55	1.3	3.202	4.765E-07	2.46E-03	0.00843	8.78E-03

where ΔT_{pv} is the change in transmittance between the valley transmittance T_v and peak transmittance T_p calculated from the experimental part of the closed aperture Zscan, as shown in fig. (6) .

$$\Delta T_{pv}= T_p - T_V \qquad \text{......................(8)}$$

$\Delta\Phi_0$ is the induced phase shift on the axis which was calculated from the equation:

$$T_{PV} = 0.406\ \Delta\Phi_0 \qquad \text{..................... (9)}$$

The nonlinear refractive index (n_2) was calculated from the equation:

$$n_2 = \Delta\Phi_{\circ} / k\, I_o\, L_{eff}, \qquad \text{................(10)}$$

where k is the wave number = $2\pi/\lambda$ and λ is the wavelength of the beam. L_{eff} is the effective length of the material, which was determined from:

$$L\ eff= (1-e^{-\alpha o L}) / \alpha_o" \qquad \text{................(11)}$$

A real part of the third-order nonlinear optical susceptibility ($Re\chi^3$) was calculated from:

$$Re\ \chi^3\ (esu) = 10^{-4}\ \varepsilon_0 c^2 n_0\ 2n_2/\pi\ (cm^2/W)" \qquad \text{...................(12)}$$

$$|\chi^3| = \left[(\mathrm{Re}(\chi^3))^2 + (\mathrm{Im}(\chi^3))^2\right]^{1/2}$$ ……………………….(13) [14, 15, 32].

T_{PV} was calculated from Figure-8, the phase shift $\Delta\Phi_0$ calculated from eq.(9), and n_2 was calculated from eq.(10), The average value of n_2 was 2.1716E^{-06} cm^2/ watt.
Figure-8 shows the nonlinear refractive index value under different initial intensities. With the increase in the laser intensity, the value of n_2 decreases. Our results have a good agreement with those of previous works [30, 31].

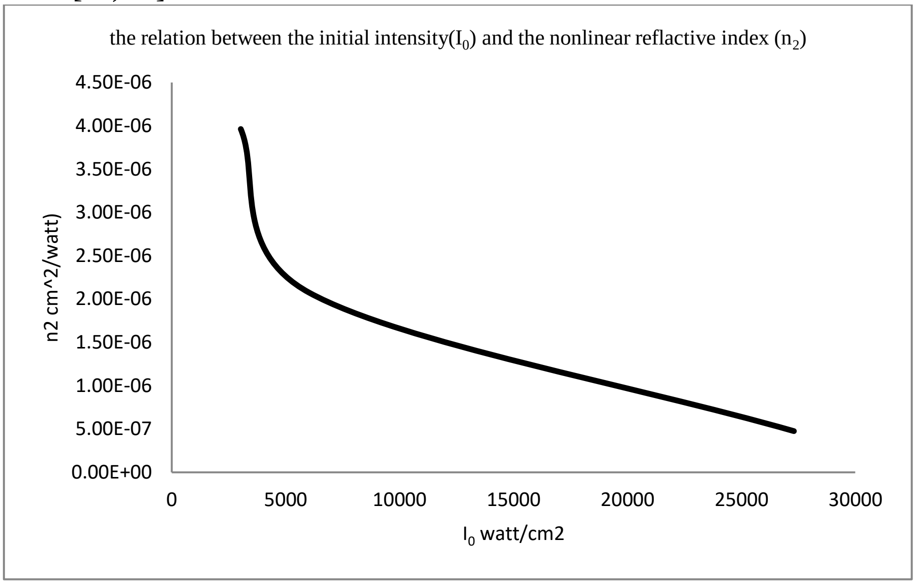


Figure 9-The relation between the nonlinear refractive index (n_2) and the initial intensity (I_0)

Conclusions

The values of n_2, β_2 and χ^3 of DQOCI doped with PMMA film were measured by using open and closed aperture Z-scan and showed intensity dependence. By changing the focal length of the lens which is used to achieve the Gaussian profile, the initial laser intensity was led to changing the above parameters. With the open aperture, z-scan curves of I_0= 3.03 and 6.38, kWatt/cm^2 showed an upward peak, indicating the occurrence of SA, in which β_2 decreased and the transmittance increased with increasing the initial laser intensity. Thus, for low intensity, this sample has useful applications in optical switching and is well suited for passive Q-switching or mode locking of laser. When I_0 was increased to 27.304 Kwatt/cm^2, nonlinear absorption was changed from SA to RSA (TPA), where the transmittances was reduced with the increase of intensity (z□0), as analyzed by the theory of free carrier nonlinearities. Also, with high intensity, the sample can be used as efficient optical limiters which can be exploited for the protection of eyes, light-sensitive sensors and CCD cameras against radiation induced damage. Closed aperture z-scan showed a pre-focal transmittance minimum (valley) and a post focal transmittance maximum (peak), which reflects the z-scan signature of a positive nonlinearity (self-focusing). This behavior is attributed to the variation of refractive index with temperature, resulting in the typical shape of a *Z*-scan trace for Kerr nonlinearity. From these conclusions, we can estimate that this kind of film will have useful applications in optical switching and is well suited for passive Q-switching or mode locking of laser.

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