

## Linear and Nonlinear Optical Properties of Acridine Dye Doped PMMA Polymer

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### ABSTRACT

*The single beam Z-scan technique was used to determine the nonlinear optical properties of the Acridine dye doped polymer film. The experiments were performed using Q-switched Nd: YAG laser with 532 nm wavelength. 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 closed aperture Z-scan data indicates that the sign of the refraction nonlinearity is negative, i.e. self-defocusing while the open aperture Z-scan data indicates that tow photon absorption coefficient. All the nonlinear optical parameters are linearly dependent with concentration.*

**Keywords:** Nonlinear Optics; concentration Effect; Z-Scan Technique

### INTRODUCTION

Nonlinear optics is the study of phenomena that occur as consequence of the modification of the optical properties of materials on interaction with intense light. Nonlinear phenomena has been studied extensively [1]. When one or more electromagnetic waves propagate in a material, the atoms and molecules oscillate not only at the frequencies of the electric field applied, but also at different combinations of those frequencies as a result of the nonlinear response of the medium. The particles of the medium are displaced from their equilibrium positions, so that positive charged particles move in the direction of the electric field, while the negative charged particles move in the direction opposite to the direction of the applied electric fields. Dipole moments are created because of the displacement between positive and negative charged particles, and the dipole moment per unit volume describes the induced polarization of the medium. When the applied electric fields are sufficiently small, the electric polarization is approximately linearly proportional with the applied electric field E

$$\mathbf{P} = \chi \cdot \mathbf{E} \dots\dots\dots (1)$$

Where  $\chi$  is the electric susceptibility tensor. This is the case of linear optics.

However, when the applied electric fields are high enough, the induced polarization has a nonlinear dependence on these electric fields and can be expressed as a power series with respect to the electric field:

$$\mathbf{P} = \chi^{(1)} \cdot \mathbf{E} + \chi^{(2)} \cdot \mathbf{E}\mathbf{E} + \chi^{(3)} \cdot \mathbf{E}\mathbf{E}\mathbf{E} \dots\dots\dots (2)$$

$$\mathbf{P} = \mathbf{P}^{(1)} + \mathbf{P}^{(2)} + \mathbf{P}^{(3)} \dots\dots\dots (3)$$

Where  $\chi^{(1)}$  is the linear susceptibility,  $\chi^{(2)}$  is the second order nonlinear susceptibility, and  $\chi^{(3)}$  is the third order nonlinear susceptibility [2].

Acridine dye is used as active medium for dye lasers, its molecular formula is  $C_{13}H_9N$  and molecular weight is  $179.22 \text{ g mol}^{-1}$  also have melting point  $107^\circ \text{C}$  while it has absorption maximum at  $390 \text{ nm}$  [3]. It is also known by the names of dibenzopyridine, 2, 3, 5, 6-dibenzopyridine and 10-azaanthracene. Acridine has an irritating odor [4]. The structure of Acridine dye is shown in Figure (1).

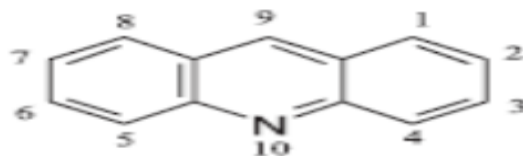


Figure 1. Acridine Dye Structure [3]

## EXPERIMENTAL WORK

Dye doped polymer films were fabricated by casting block method, the concentrations are ( $1 \times 10^{-3}$ ,  $5 \times 10^{-4}$ ,  $1 \times 10^{-4}$  and  $5 \times 10^{-5}$ ) Ml. The solution of the polymer is prepared by dissolving the required amount of polymer (7 gm in 100 ml of chloroform solvent). A desired amount of dye solution was added to polymer solution and stirred by a magnetic stirrer at room temperature to get a uniform mixture. Films were shaped by drying the mixed polymer dye solution on a glass block at room temperature  $25^\circ \text{C}$ . Flow diagram of fabrication dye doped polymer film is shown in Figure (2).

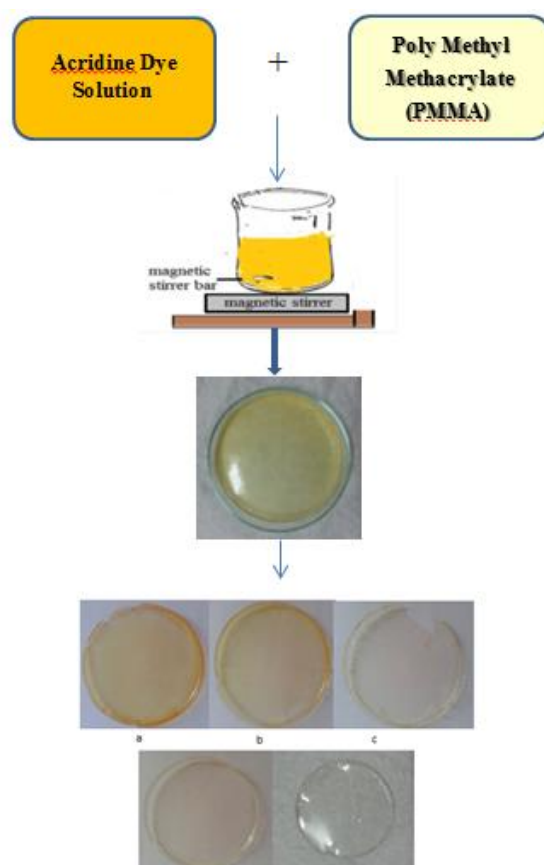


Figure 2. Acridine Dye Doped PMMA at Different Concentrations (a =  $1 \times 10^{-3}$ , b =  $5 \times 10^{-4}$ , c =  $1 \times 10^{-4}$ , d =  $5 \times 10^{-5}$ , and e = PMMA)Ml

Figure (3) shows the experimental z-scan set up. It consists of a 30 ns Q-switched Nd: YAG laser operating at  $1064 \text{ nm}$  wavelength with

energy of 20 mJ. Laser pulse energy was measured by the (DPSS 1830C) detector. The laser beam passes through a lens of 10 cm focal length. Sample was moved through the beam waist of laser beam along the z axis distance using a translating stage.

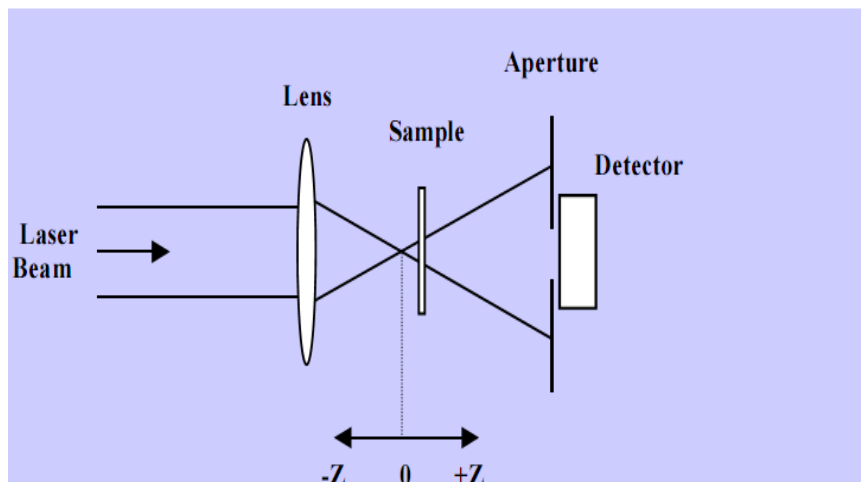


Figure 3. Experimental set-up for Z-scan

## RESULTS AND DISCUSSION

### The Linear Optical Properties

#### Absorption Spectra

The absorption spectra of Acridine dye at the concentrations ( $5 \times 10^{-5}$ ,  $1 \times 10^{-4}$ ,  $5 \times 10^{-4}$  and  $1 \times 10^{-3}$ ) Ml after doped by PMMA polymer was shown in Figure (4).

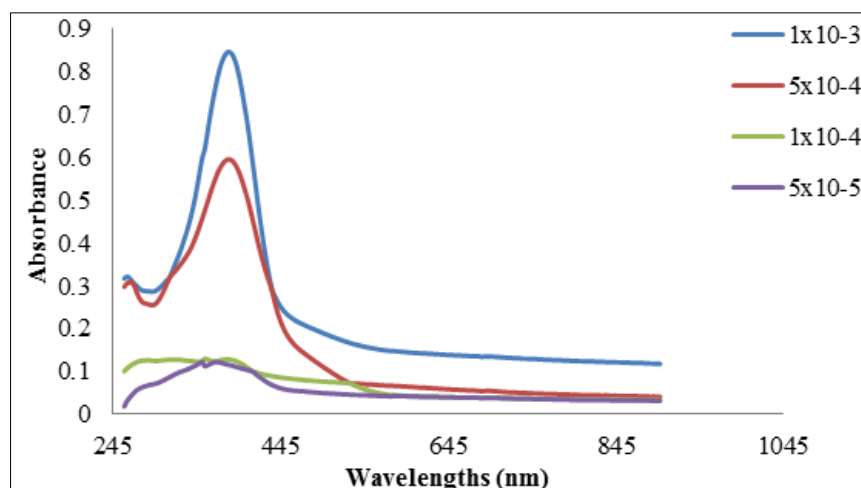


Figure 4. UV-VIS Absorption Spectrum of the Acridine Dye Doped PMMA Polymer

From Figure (4), absorption peaks are (376, 378, 379 and 385) nm at different concentrations ( $5 \times 10^{-5}$ ,  $1 \times 10^{-4}$ ,  $5 \times 10^{-4}$  and  $1 \times 10^{-3}$ ) Ml respectively also shifted toward the longer wavelengths with increasing concentrations. This shift was overall obtained due to the increasing number of molecules per volume unit at high concentration, this in turn lead to change in energy levels result in effect of vibration field on molecules. Therefore, absorbance increased with increasing concentration.

**Transmission Spectra**

The transmission spectra of Acridine dye at concentrations ( $5 \times 10^{-5}$ ,  $1 \times 10^{-4}$ ,  $5 \times 10^{-4}$  and  $1 \times 10^{-3}$ ) Ml after doped PMMA polymer were shown in Figure (5).

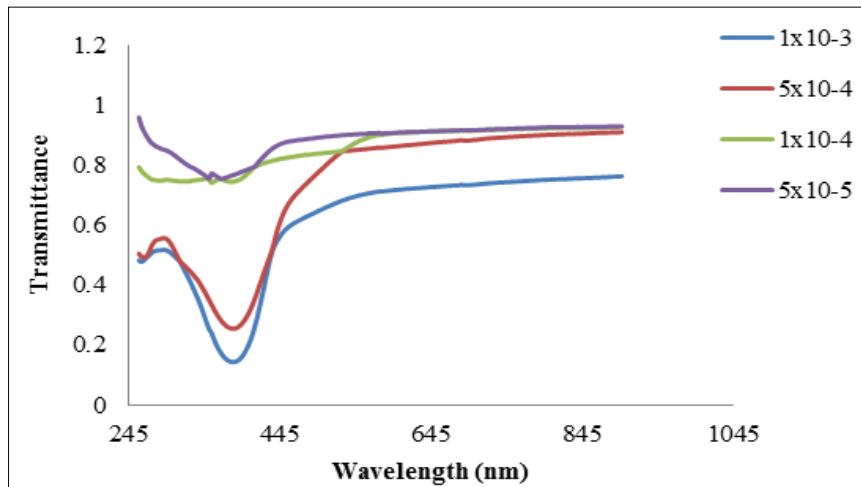


Figure 5. UV-VIS Transmittance Spectrum of the PMMA after the addition of different concentrations of Acridine Dye

Transmission values at 532 nm are (80,77,73 and 68)% at ( $5 \times 10^{-5}$ ,  $1 \times 10^{-4}$ ,  $5 \times 10^{-4}$  and  $1 \times 10^{-3}$ ) Ml respectively. Transmission values in this case decreased with increasing concentration.

The linear absorption coefficient ( $\alpha_o$ ) and linear refractive index ( $n_o$ ) obtained from eq. (4), eq. (5). The linear refractive index and linear absorption coefficient of acridine dye listed in Table (1) [5].

$$\alpha_o = \frac{1}{t} \ln\left(\frac{1}{T}\right) \dots \dots \dots (4)$$

$$n_o = \frac{1}{T} + \left[\left(\frac{1}{T^2} - 1\right)\right]^{1/2} \dots \dots \dots (5)$$

Where  $t$  is the thickness of sample, and  $T$  is transmission Acridine dye doped PMMA at 532nm, the linear absorption coefficient ( $\alpha_o$ ) and linear refractive index ( $n_o$ ) were determined for Acridine dye doped PMMA at 532nm. These values are summarized in Table (1).

**Table 1. Linear Absorption Coefficient and Linear Refractive Index of Acridine Dye Doped PMMA**

Concentration Ml	$\alpha \text{ cm}^{-1}$	$n_o$
$5 \times 10^{-5}$	13.9	2
$1 \times 10^{-4}$	15.6	2.11
$5 \times 10^{-4}$	18.5	2.3
$1 \times 10^{-3}$	22.2	2.54

## The Nonlinear Optical Properties

### The Nonlinear Refractive Index

Acridine dye solution doped PMMA polymer at the concentrations ( $5 \times 10^{-5}$ ,  $1 \times 10^{-4}$ ,  $5 \times 10^{-4}$  and  $1 \times 10^{-3}$ ) Ml, the closed aperture measurements were shown in Figure (6) at wavelength 532 nm.

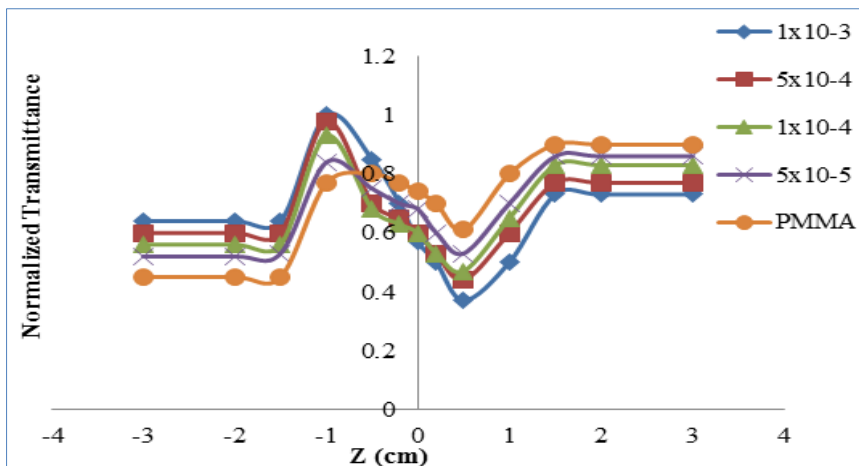


Figure 6. Closed-Aperture of Acridine Dye Doped PMMA at different Concentrations

In Figure (6), the nonlinear region was extended from -1.5 cm to 1.5 cm and the transmittance difference between peak and valley,  $\Delta_{p-v}$  at concentrations ( $5 \times 10^{-5}$ ,  $1 \times 10^{-4}$ ,  $5 \times 10^{-4}$  and  $1 \times 10^{-3}$ ) Ml equals to (0.31, 0.46, 0.54 and 0.63) respectively. Also high transmission was 1 and small transmission was 0.37 at concentration  $1 \times 10^{-3}$  Ml. Acridine dye doped PMMA polymer indicates that the sign of the refraction nonlinearity is negative, i.e. self-defocusing observed in this figure

In order to describe the z-scan behavior in the previous figures. When the sample moves far from the focus, the transmitted beam intensity is low and the transmittance remains relatively constant. As the sample approaches the beam focus, intensity increases, leading to self-lensing in the sample tend to collimate the beam on the aperture in the far field, increasing the measured transmittance at the iris position. If the beam experiences any nonlinear phase shift due to the sample as it is translated through the focal region, then the fraction of light falling on the detector will vary due to the self-lensing generated in the material by the intense laser beam. In this case, the signal measured by detector will exhibit a peak and valley as the sample is translated. The position of the peak and valley, relative to the z-axis, depends on the sign of the nonlinear phase shift. Where the change in the normalized transmittance from the peak of the curve to the valley ( $\Delta T_{pv}$ ) is directly proportional to the nonlinear phase shift imparted on the beam. Moreover, if the beam is transmitted through the nonlinear medium the induced phase shift can also be either negative or positive accordingly when the medium is self-defocusing or self-focusing, respectively. The magnitude of the phase shift can be determined from the change in transmittance between peak and valley.

After the focal plane, the self-defocusing increases the beam divergence, leading to a widening of the beam at the iris and thus reducing the measured transmittance. Far from focus ( $z > 0$ ), again the nonlinear refraction is low resulting in a transmittance z-independent [6]. The nonlinear refractive indices were determined from equation (6) at 532 nm was shown in Tables (2).

$$n_2 = \Delta\Phi_0 / I_0 L_{eff} k \dots\dots\dots (6)$$

Where  $\Delta\Phi_0$ : nonlinear phase shift

$$\Delta\Phi_0 = \Delta T / 0.406 \dots\dots\dots (7)$$

$k = 2\pi / \lambda$ ;  $\lambda$ : is the wavelength of the beam.

$I_0$  is the intensity at the focal spot given by

$$I_0 = P_{\text{peak}} / \pi\omega^2 \dots\dots\dots (8)$$

$\omega$ : the beam radius at the focal point,

$P_{\text{peak}}$ : the peak power given by

$$P_{\text{peak}} = E / \Delta t \dots\dots\dots (9)$$

Where

E: The energy of the pulse

$\Delta t$ : The pulse duration.

$L_{\text{eff}}$ : The effective length of the sample can be determined from the following formula.

$$L_{\text{eff}} = (1 - e^{-\alpha_0 L}) / \alpha_0 \dots\dots\dots (10)$$

Where,

L: the sample length,

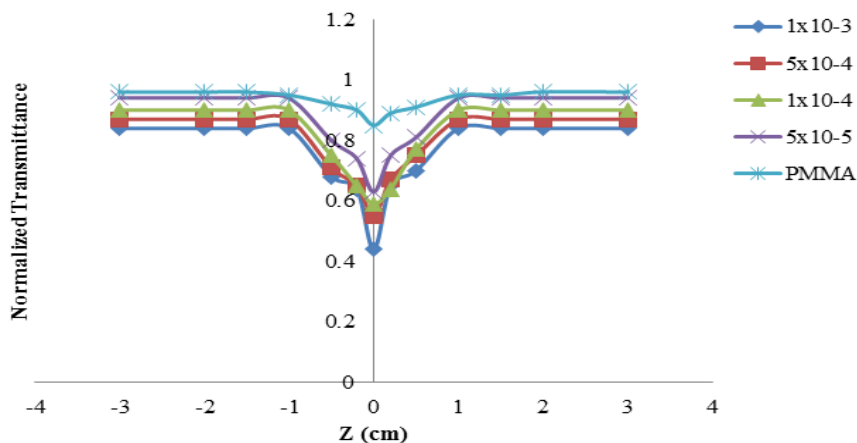
$\alpha_0$ : linear absorption coefficient [7].

**Table 2. Nonlinear Refractive Index for Acridine Dye Doped PMMA at different Concentrations**

Concentrations (Ml)	$n_2, \text{cm}^2/\text{GW}$
$5 \times 10^{-5}$	0.0755
$1 \times 10^{-4}$	0.1073
$5 \times 10^{-4}$	0.12381
$1 \times 10^{-3}$	0.1419

**Nonlinear Absorption Coefficient ( $\beta$ )**

To investigate the nonlinear absorption coefficient, at wavelength 532nm. Figure (7) shows open-aperture z-scan at different concentrations at 532nm.



**Figure 7. Open-Aperture Z-Scan of Acridine Dye Doped PMMA at different Concentrations**

Figure (7), showed the valley is larger because of effecting PMMA polymer and  $T_{min}$  are (0.63, 0.59, 0.55 and 0.44) at the concentrations ( $5 \times 10^{-5}$ ,  $1 \times 10^{-4}$ ,  $5 \times 10^{-4}$  and  $1 \times 10^{-3}$ ) Ml.

The behavior of transmittance started linearly at different distances from the far field of the sample position (-Z). At the near field the transmittance curve begins to decrease until it reaches the minimum value ( $T_{min}$ ) at the focal point, where  $Z=0$  mm. Afterward, the transmittance begins to increase toward the linear behavior at the far field of the sample position (+Z).

The nonlinear absorption coefficient at 532nm were determined from equation (11) was shown in Tables (3).

$$T(z) = \sum_{m=0}^{\infty} \left[ \frac{\beta I_0 L_{eff}}{1 + (Z/Z_0)^2} \right]^m \dots\dots\dots (11)$$

Where,

Z: is the sample position at the minimum transmittance; m: integer.

T(z): the minimum transmittance [7].

**Table 3. Nonlinear Absorption Coefficients at different Concentrations of Acridine Dye Doped PMMA**

Concentrations (Ml)	B (cm/GW)
$5 \times 10^{-5}$	41948.3
$1 \times 10^{-4}$	38271.2
$5 \times 10^{-4}$	35688.9
$1 \times 10^{-3}$	29781.5

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