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Non-Linear Optical Properties of Organic Laser Dye

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Abstract

The present research includesstudy spectral, linear and nonlinear optical properties of (Acriflavine)organic laser dye at concentrations (10^{-5}M),and preparing thin films of this dye doped with polymer and (Ag) nano material, for using it in field of nonlinear optics. Many properties of organic dyes can be improved and enhanced by doping with polymer and nanoparticles materials, which are favorable for practical applications. The nonlinear calculated included using (Z - Scan) technique.The measurements performed using diode pump solid state laser operating at (457nm) wavelength with different laser power (56,70,84 and 102 mW). The results showed that increasing the nonlinear refractive index when increasing powers but decreasing of nonlinear absorption coefficient, when increasing powers for all prepared samples. Thin films of dye doped with the polymer and Ag nano material have shown better nonlinear properties and optical limiting as compared with samples of pure dye.The result imply that all samples can be used as a potential medium for various optoelectronic applications including that in optical power limiting.

Key words: Organic laser dyes, Z-Scan technique, Optical limiting, Non-linear optical properties.

I. Introduction

The organic compounds are defined as hydrocarbons and their derivatives. They can be subdivided into saturated and unsaturated compounds. Organic material can show very high nonlinear coefficients, because of the large variety of these compounds at high intensities. Nonlinear optical (NLO) properties have been the subject of numerous investigations theoretically and experimentally during recent years due to their applications to many branches. The nonlinear optical properties are important parameters in characterizing and determining the applicability of any material to nonlinear optical device[1]. Nonlinear optical parameters can be studied by using a simple technique called Z- Scan. It is a sensitive and popular experimental method to measure intensity dependent of non-linear optical properties of materials which can rapidly measure both non-linear absorption (NLA) and nonlinear refraction (NLR) in solids, liquids and then studying the optical limiting behavior [2].

By using the Z-Scan technique the optical nonlinearity of many organic materials has been explored. The Z-Scan technique is a simple and sensitive method introduced in 1990 to measure the nonlinear refractive index of optical materials. In this technique the sample is moved in the intensity pattern generated along the z-axis by a focused laser beam, and the on-axis transmitted signal is measured in the far field by a detector placed behind a small aperture change of the refractive index due to optical nonlinearities in the sample is detected as characteristic intensity variation of the transmitted signal as a function of the sample position. The Z-Scan setup has been used to detect small linear absorption using low power continuous wave (CW) lasers.

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Optical limiting with organic materials or devices with transmission that decreases with light level is called optical limiters and has applications in eye and sensor protection. Ideally, an optical limiter should exhibit linear transmittance at low incident light, but become opaque at high incident light [3].The organic compounds are defined as hydrocarbons and their derivatives. They can be subdivided into saturated and unsaturated compounds [4].Organic laser dyes are fluorescent particles with large molecular weights, portrayed by containing broadened systems of conjugated double bonds. In a dye laser, these molecules are dissolved in an organic solvent. Usually, they have a good absorption band [5]. Laser dyes are perplexing molecules containing various ring structures, which lead to complex absorption and emission spectra. It can be sorted into various classes by virtue of their structures that are chemically comparative. Basic examples are the coumarin, xanthene and pyrromethene. The structure and arrangement of the molecules has an imperative impact on spectral emission [6]. The aim of this work was the use of the Z-Scan technique to study the nonlinear optical properties of (Acriflavine)organic laser dye at concentrations (10^{-5}M)as solution and thin films with (CW) diode solid state laser at (λ =457 nm) wavelength and (56,70,84,102mW) power, for using it as a good limiter device for many applications.

II. Theory

There is considerable interest in finding materials having fast nonlinearities. This interest, that is driven primarily by the search for materials for all-optical switching and sensor protection applications, concerns both nonlinear absorption (NLA) and nonlinear refraction (NLR). At high intensity, the absorption of the substance is provided by [7]:

Where Iis the incident intensity, α_oIt is the coefficient of linear absorption and β is the intensity-related nonlinear absorption coefficient. The refractive index is given at high intensity by [8]:

The coefficient of linear refraction is non-linear and the coefficient of non-linear refraction is n_2. Z-Scan technique, which can be used to investigate nonlinear optical properties.The nonlinear refractive index is calculated from the peak to valley difference of the normalized transmittance by the following formula [9]:

Where $\Delta\Phi_o$is the nonlinear phase shift [10]:

ΔT_{p-v} the difference between the normalized peak and valley transmittances, $k = 2\pi/\lambda$, λ is the wavelength of the beam, Io is the intensity at the focal spot and L_{eff} is the sample's effective length, calculated from [11]:

Where L is the sample length and α_o is linear absorption coefficient which is given as [9]:

Where (t) is the thickness of sample and T is the transmittance. The linear refractive index (n_o) obtained from equation [12]:

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The intensity at the focal spot is given by [10]:

Is defined as the peak intensity within the sample at the focus, where ω_o is the beam radius at the focal point. The coefficients of nonlinear absorption (β), can be easily calculated by using following equation [12]:

Where T(z): The minimum value of normalized transmittance at the focal point, where (Z=0).

III. Materials used

a-AcriflavineOrganic Dye

Acriflavine is derived from (acridine family). Figure (1) shows the chemical structure of this dye.Table (1) shows the properties of the Acriflavine dye.

Table (1) Properties of the Acriflavine dye.

Chemical Name	Molecular Formula	Molecular Weight
Chromoflavine	$C_{14}H_{14}CLN_3$	259.737 g/mol

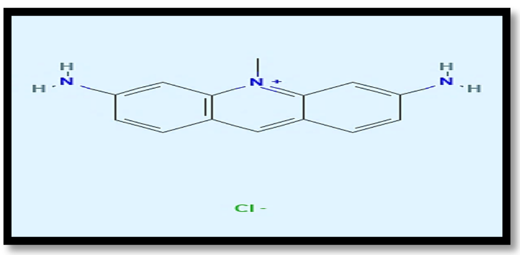

Figure (1):molecular structure of Acriflavineorganic dye.

b- Ethanol Solvent

Ethanol also called (ethyl alcohol) is a pure alcohol, colorless liquid. Ethanol can cause alcohol intoxication when consumed. It burns with blue flame. The physical properties of ethanol stem primarily from the presence of its hydroxyl group and the shortness of its carbon chain. Volatile colorless liquid and its density (0.789 gm / cm^3) and molecular weight (46.07 gm / mol).

c-Polymer

Polyvinyl alcohol (PVA) was utilized in present work; the properties of this polymer are listed in table (2).

Table (2): The principal qualities of polymer.

Molecular formula	Avantor (Netherlands)

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Solubility	Water soluble
Physical Appearance	White, odorless

d- Silver nanoparticles (Ag NP_S)

Silver nanoparticles were supplied by laboratory Reagent LTD; it has main characteristics are shown in Table (3).

Table (3): The main characteristics of Ag NPs nanoparticles

Ag nanoparticle	Description
Productive company	SIGMA-ALDRICH, Germany
Molecular formula	Ag NPs
form	Nano powder
Surface area	> 40 m^2/g
Particle size	(1-100) nm

E. Samples Preparation

Solutions of concentrations (10^{-5}) M of (Acriflavine)at(10^{-5}M) concentration, dissolve (0.0029gm) of the dye powder in the size of (10 cm^3) of the ethanol solventthe powder was weighted by using an electronic balance type (BL 210 S), Germany, having a sensitivity of four digits according to the equation.

$$W = \frac{M_W \times V \times C}{1000} \qquad (10)$$

Where, W: Weight of the dissolved in material (g), Mw: Molecular weight of the material (g /mol), V: Volume of the solvent (mL) and C: The concentration (M).
To prepare lighter concentration (10^{-5}M) From the concentration prepared, the following relationship, called the dilution relationship, is used [12]

$$C_1V_1 = C_2V_2 \qquad (11)$$

Where: C_1: Primary concentration, C_2: New concentration. V_1: The volume before dilution and V_2: The volume after dilution.

The thin films of Acriflavine, were prepared on a clean glass slide via drop casting method, with solution at concentration (10^{-3}) M for each of them, and dried at room temperature for (3) days, the thickness of these thin films is about (150-250) nm. Michelson's interferometer was utilized to measure the thickness of dye doped thinfilm. In the Michelson's interferometer, the collimated beam of light is divided into two parts by partial reflection, which can be used to measure exceedingly small distances in terms of the wavelength of light. Dye doped polymer films were fabricated by drop casting method, at concentration (10^{-3}) M. The solution of the polymer is prepared by dissolving the required amount of polymer (2 mg in 50 mL of Ethanol solvent). was utilized to measure the thickness of dye doped thinfilm [13].

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IV. Result and Discussion

A-Linear Optical Coefficients

(Acriflavine) optical measurements including spectral transmittance (T) and absorbance (A) obtained by means of a spectrophotometer (Shimadzu UV-VIS) with a wavelength range between (190-1100) nm. The survey of all wavelengths is carried out by a computer programmer and the wavelength at which maximum absorption occurs and less transmittance is given. Thin films at (10^{-3} M) were prepared by drop casting method at room temperature. The linear absorption spectra of thin films of (Acriflavine) at 10^{-3} M were shown in Figures (2). The present results show that the absorption peaks for (PVA and Nano) at 10^{-3} M in Ethanol solvent were shifted toward the longerwavelengths after addition (PVA) polymer and nanoparticles to pure dye.

The transmission spectra of thin films of (Acriflavine, PVA, Nano), at 10^{-3} M, as shown in Figure (3). The optical transmission curve of all samples showed a variable behavior of the transmission as a function of the incident wavelength. The transmittance intensity of Nano composite was lower than intensity (dye without adding and dye doped with PVA). Transmittance decreases, this is suggested to take place because of Ag NP_S nanoparticle contain electrons which can be the absorption of electromagnetic energy of the incident light and travel to higher energy levels. This process is not accompanied by emission of radiation because the traveled electron to higher levels have occupied vacant positions of energy bands, thus part of the incident light is absorbed by the substance and does not penetrate through it.

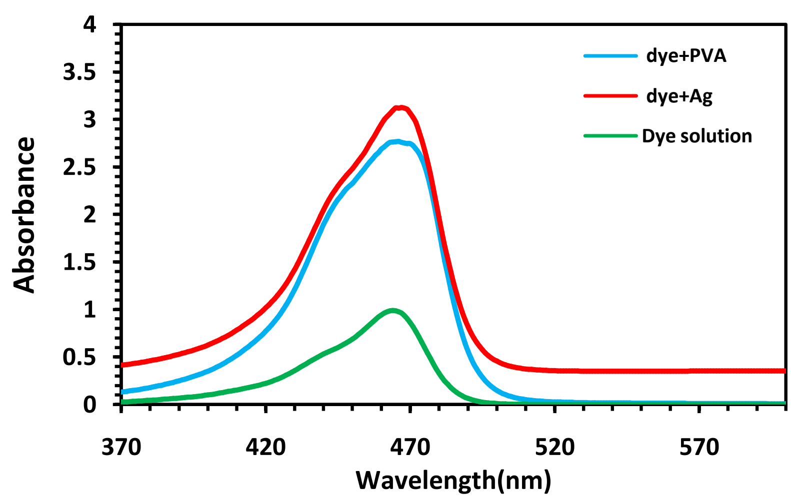

Figure (2): The absorption spectra of Acriflavine dye as: (solution dye,dye +PVA,dye+PVA+ Nano).

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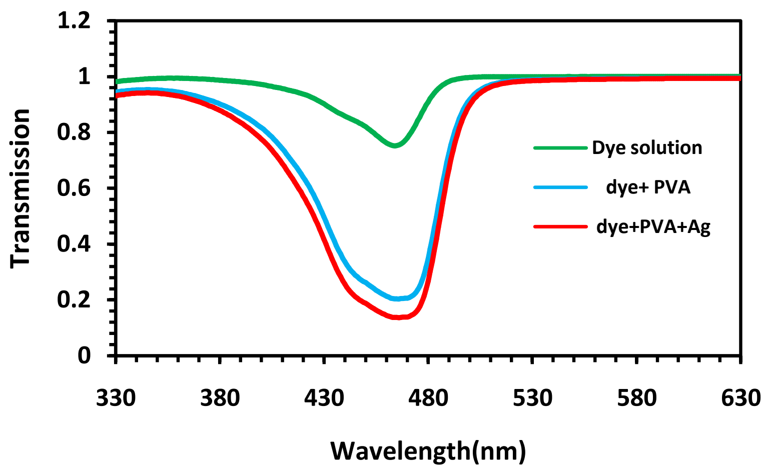

Figure (3): Transmission spectra for of Acriflavine dye as: (solution dye, dye +PVA,dye +PVA+ Nano).

B. Z-scan technique

The non-linear refractive index (n_2) and the coefficient of non-linear absorption of all samples are calculated using the z-scan process. Using a continuous wave diode pump solid state laser at (λ =457 nm) as the light source, we used the closed aperture Z-scan process. A CW laser Gaussian beam with a well-defined vertical polarization and the strength of the laser (56,70,84,102 mW).To focus the laser beam on the sample, the convex lens with a focal length of (15 cm) was used; the samples were excited for the laser beam at normal incidence geometry. The sample was shifted back and forth along the z-axis around the minimum beam waist of the laser during the Z-scan calculation in order to measure transmittance as a function of the sample location. The configuration diagram for the z-scan experiment is shown in Fig (4).

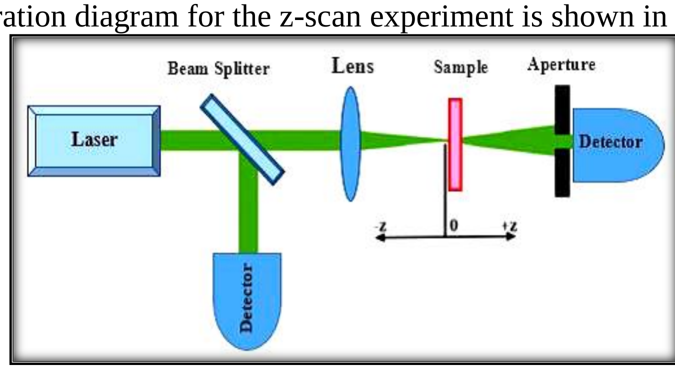

Figure (4): Schematic diagram of the experimental setup for closed aperture z-scan technique.

C.Nonlinear optical properties

The nonlinear absorption coefficientof investigated for (Acriflavine) for different powers (56,70,84,102) mW in Ethanol solvent were measured by open-aperture Z-Scan technique. The performed open aperture Z-Scan exhibits an increasing in the transmission about the focus of the lens. Open-aperture Z-Scan of samples at (457nm), in (Ethanol) solvent are shown in Figures (5).

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Its noticed (two photon absorption) phenomenon.The behavior of transmittance starts 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). The transmittance begins to increase towards the linear behavior at the far field of the sample position (+Z). The change of intensity, in this case, is caused by two photon absorptions when in the sample travels through beam waist. The open-aperture Z-Scan defines variable transmittance values, which was used to determine absorption coefficient.In the focal plane where the intensity is greatest, the largest nonlinear absorption is observed.

Saturable Absorptionphenomenon were observed for open-aperture Z-Scan of thin films of compounds under study as shown in Figures (6 and 7). The behavior of transmittance curves starts linearly at different distance from the far field of the sample position (-Z). At the near field the transmittance curve begins to increase until it reaches the maximum value (T_{max}) at the focal point, where Z=0 mm. Afterwards, the transmittance begins to decrease toward the linear behavior at the far field of the sample position (+Z). The transmittance is sensitive to the nonlinear absorption as a function of input power intensity. The change in intensity is caused by saturation absorption in the sample as it travels through the beam waist. In the focal plane where the intensity is greatest, the largest nonlinear absorption is observed. At the far field of the Gaussian beam, where, the beam intensity is too weak to elicit nonlinear effects. A symmetric peak value is contributed to the negative nonlinear absorption coefficient β, indicates that the sample shows a bleaching-like behavior (saturation of absorption).

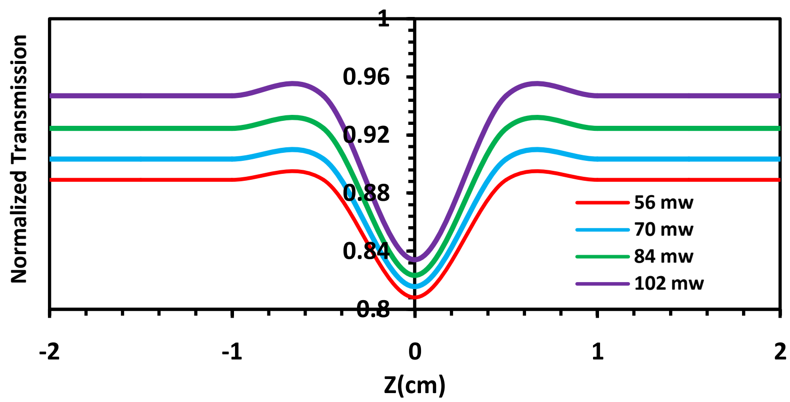

Figure (5): Open-aperture Z-Scan data of Acriflavineas solution with different laser power

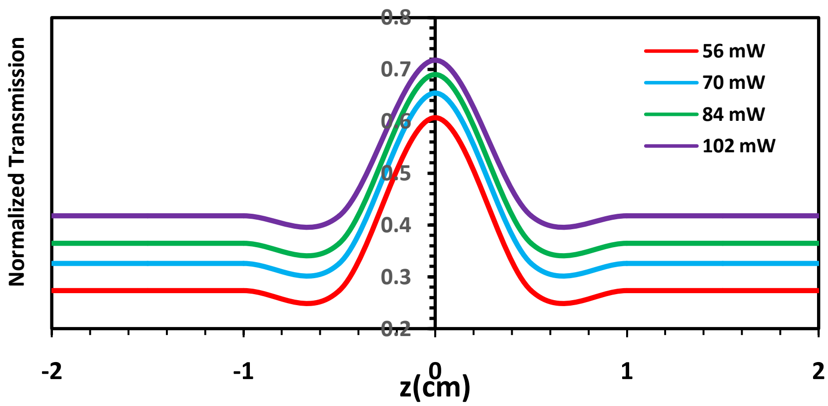

Figure (6): Open-aperture Z-Scan data for thin film of (Acriflavine +PVA)

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with different laser power

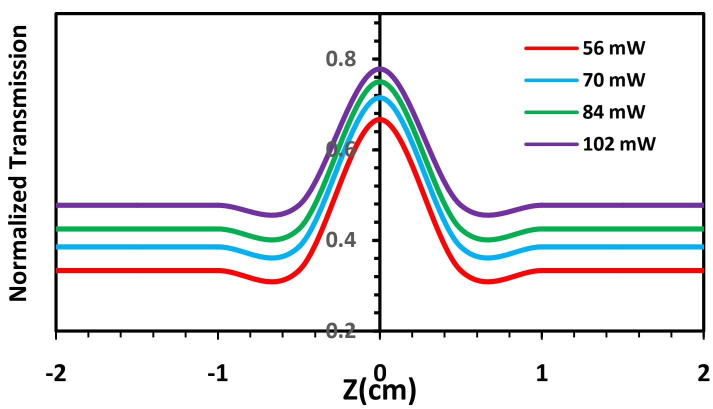

Figure (7): Open-aperture Z-Scan data for thin film of (Acriflavine +PVA+(Ag)nano material)
with different laser power

D. Nonlinear Refractive Index

The nonlinear refractive index of the prepared dyein concentration (10^{-5}) M in Ethanol solvent were measured by closed-aperture Z-Scan technique. The normalized transmittances of Z-Scan measurements as a function of distance in (Ethanol) solvent is shown in figure (8) the nonlinear effect region is extended from (-1.5) mm to (1.5) mm the nonlinear medium acts as a positive lens so that it focuses the transmitted beam through the hole, causing an increase in the intensity transmitted from the sample to the detector, and when the sample moves towards a decrease in intensity, the effect of the optical medium is similar to the effect of a negative lens that disperses the transmitted beam away from the aperture, which leads to a decrease in the intensity transmitted from the sample to the detector,
From the results in figures (9 and10) the closed-aperture z-scan measures the transmittance of a sample, as it passes through the focal plane. The sample nonlinearity was calculated from the difference between the heights (peak) and the lowest value (valley) transmission values denoted as ΔT_{p-v}. The valley in thetransmission curves emerge first, and then the peak appears after the sample through the waist. This is referring to a positive nonlinear refractive index. This dependence can be explained as follows. The sample acts as a focusing lens (thermal lens), and for negative z the enlarged beam waist is shiftedtowards the positive z coordinate. Therefore, the beam width beside the aperture becomes larger and transmittance decreases. At the waist, the sample had also no effect because the curvature radius is infinite in this position. After the sample passes through the waist and moves toward the aperture.The nonlinearity of doped dye is larger than those for solution dye. The nonlinear parameters are calculated, as tabulated in Tables (4) from this Table we show that the values of nonlinear parameter for(n_2) are increase with increase the powers and (β) decrease when the powers increase. The closed-aperture Z-Scan defines variable transmittance values, which used to determine the nonlinear phase shift $\Delta\Phi_o$.

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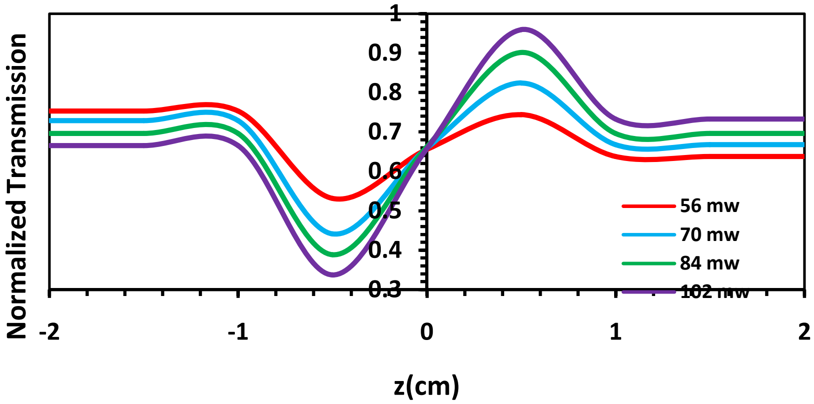

Figure (8): Closed-aperture Z-Scan data of Acriflavineas solution with different laser power

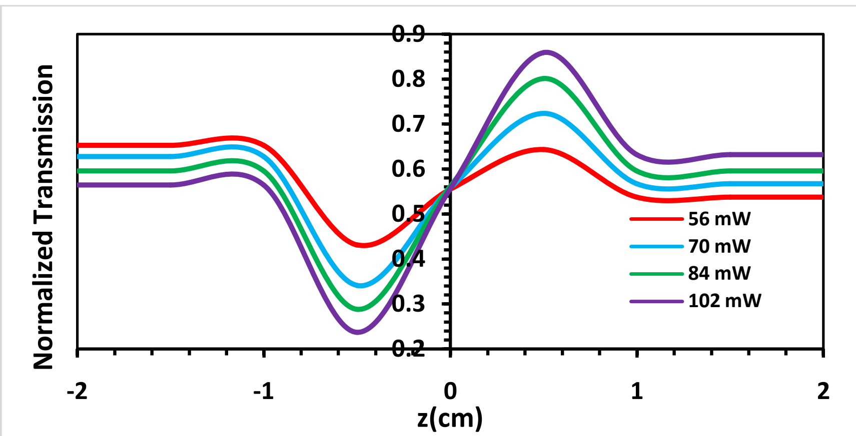

Figure (9): Closed-aperture Z-Scan data for thin film of (Acriflavine +PVA) with different laser power

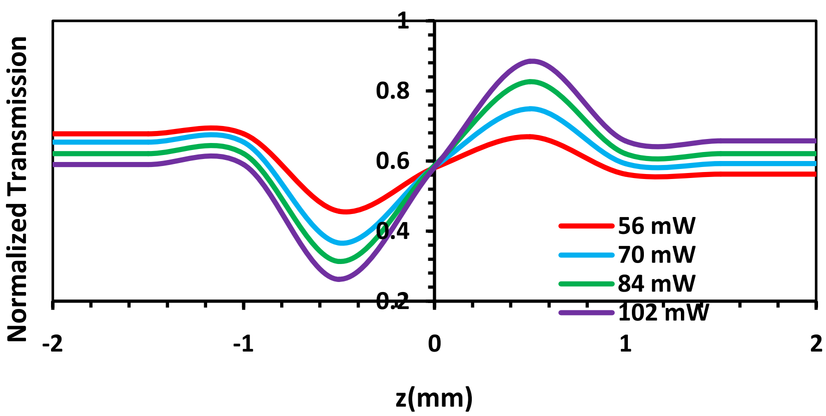

Figure (10): Closed-aperture Z-Scan data for thin film of (Acriflavine +PVA+ +(Ag) nano material with different laser power.

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Table (2): The nonlinear optical parameters of Acriflavineat λ=457nm with different laser power

Material	Power (mW)	ΔT_{P-V}	$\Delta\Phi_o$	n_2 (cm^2/mW)	T (z)	β (cm/mW)
Acriflavine	56	0.2123	0.5231	6.7541×10^{-10}	0.8081	0.401×10^{-3}
	70	0.3826	0.9423	9.7338×10^{-10}	0.8156	0.324×10^{-3}
	84	0.5128	1.2631	10.0872×10^{-10}	0.8234	0.272×10^{-3}
	102	0.6218	1.5316	10.1657×10^{-10}	0.8342	0.244×10^{-3}
Acriflavine + PVA	56	0.2623	0.6462	5.0697×10^{-6}	0.6066	2.0002
	70	0.4325	1.0655	6.6868×10^{-6}	0.6542	1.7257
	84	0.5628	1.3862	7.2498×10^{-6}	0.6900	1.5166
	102	0.6718	1.6548	7.6521×10^{-6}	0.7177	1.3950
Acriflavine + PVA+Nano	56	0.2923	0.72015	6.1090×10^{-6}	0.6566	3.5650
	70	0.4625	1.1393	7.7323×10^{-6}	0.7042	2.1576
	84	0.5928	1.4601	8.2575×10^{-6}	0.7412	1.8292
	102	0.7018	1.7286	8.6442×10^{-6}	0.7677	1.6922

E. Optical Limiting Behavior

The optical limiting behavior was conducted with the same laser closed-aperture using Z-Scan technique.Figure (10) provides the optical limiting characteristics for the sample. Initially, the output power increases with the increase in input power, but the samplesbegin defocusing the beam after a certain threshold.Thin films of Acriflavine give optical power limiting threshold and limiting amplitude less as compared with the materials as solutions, therefore, the optical limiting behavior was significantly optimized in the case of thin films in comparison to liquid specimens and in the polymer and nanoparticles modified dyes in general.

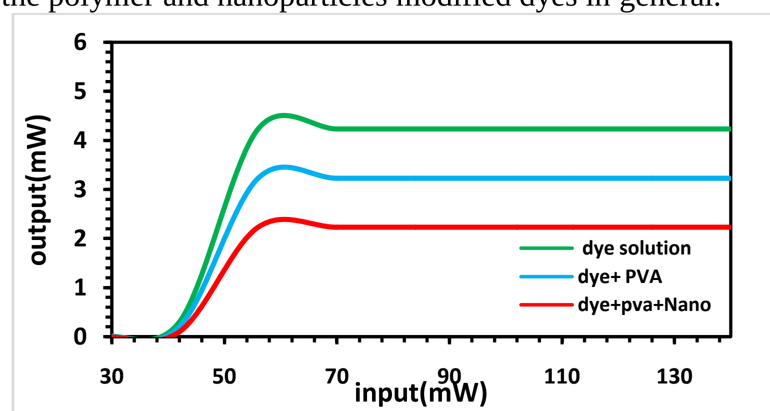

Figure (10) Optical limiting ofAcriflavinedye as: (dye solution, dye +PVA,

dye +PVA+Ag Nano) .

V. **Conclusions**

The non-linear refraction index and the non-linear absorption coefficient for solution of (Acriflavine) organic laser dye in ethanol solvent at (10^{-5} M) concentration and thin films of this dye doped with polymer and (Ag) nano material have been measured,using the Z-Scan technique with (457 nm) of (CW) laser with different laser power (56,70,84 and 102 mW). The results showed that increasing the nonlinear refractive index when increasing powers but decreasing of nonlinear absorption coefficient, when increasing powers for all prepared samples. Thin films of dye doped with the polymer and Ag nano material have shown better nonlinear properties and optical limiting as compared with samples of pure dye .The result imply that all samples can be used as a potential medium for various optoelectronic applications including that in optical power limiting.

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