

NONLINEAR PROPERTIES OF OLIVE OIL FILMS DOPED WITH POLY (METHYL METHACRYLATE), POLYSTYRENE AND THEIR BLEND BY USING Z- SCAN TECHNIQUE

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ABSTRACT

The nonlinear optical properties and optical limiting of olive oil (OO) films doped with PMMA, PS, and their blends at different ratios of olive oil and doped with polymers (1 /2 (50%), 1/3 (33 %), ¼ (25%)) was studied. The films deposited by free-casting technique and characterized by Z- scan technique that was performed using continuous wave (CW) diode laser at 650 nm, at two parts closed aperture for measuring third order nonlinear refractive index and open aperture to measure nonlinear absorption coefficient. The effect of nonlinear refractive index of doped films is obtained by measuring the profile of diffused beam through the samples, and their nonlinear refractive index is obtained to be positive results from self-focusing. With open aperture Z-scan, it is found that most the films exhibited saturation absorption behavior, except films of 50% OO/PMMA and 50% OO/ (PMMA+PS) changes from the saturable absorption (SA) to reverse saturable absorption (RSA) where's the ratio of olive oil is increased. The optical limiting behavior of samples is investigated by measuring the output and input; it is found that films with blend polymer have the best optical limiting behavior. Real and imaginary parts of third-order nonlinear optical susceptibility were measured using Z-scan data; it was changed significantly with the doping ratio.

I. INTRODUCTION

Olive oil (OO) represents the one class of lyotropic liquid crystal in nematic phase [1]. Liquid Crystals (LC) are characterized by large optical nonlinearities which have been studied in recent years from both the theoretical and practical considerations [2]. Since LC molecules typically have delocalized π electrons, they are believed to be potential sources of fast and large nonlinearities.

Nonlinear optical parameters such as nonlinear refractive index and nonlinear optical absorption are expected to relate with the geometrical configuration between the light polarizing direction E and the nematic director n^o [3]. Olive oil behaves as a nonlinear material where it has a "blue shift" in some excited wavelengths. For this olive oil nonlinear optical properties have prominent importance for photonics applications [1].



Nonlinear optical phenomena may be caused by electronics and nonelectronics processes [4]. The former refers to those radiative interactions between the active electron and the optical electric field. Usually, these processes are very fast, in period around picoseconds, and spatially localized, such as second and third harmonic generation. There are many techniques have been used to measure the nonlinear optical refractive index such as nonlinear interferometry, degenerate four-wave mixing and ellipse rotation. These techniques are susceptible and require complex laboratory apparatus. The Z- scan technique is simple and meticulous in measuring the optical nonlinear parameters such as nonlinear refractive index and nonlinear absorption. This technique is simple and useful to study nonlinear optical properties of materials in different time scales [5].

The optical limiting is a nonlinear optical process in which the intensity transmittance through the material decreases with increased the intensity of the incident light. The performance of optical limiting will be improved by coupling two or more mechanisms of optical nonlinearity. Reverse saturable absorption (RSA) and excited state absorption (ESA) are the most popular mechanism for the nonlinear optical behavior of organic materials [6]. The effects of optical nonlinear can be employed for the optical limiting design and its performance. Limiter has a linear transmitted intensity at low input intensities, but above the threshold intensity, its transmitted intensity becomes constant [7]. Materials with strong nonlinear optical absorption properties have been recognized as prospective optical limiting materials for applications such as pulse shaping in various wavelength regions, and sensor protection noise reduction [8]. The main requirements for optical limiting materials are large pulse energy suppression, high damage threshold, high linear transmission, and low turn-on threshold [9].

Recently, the material researchers have been used the copolymers and their blends to obtained intermediate properties that related to homo-polymers for some particular functions. These properties are related to the molecular motions in their amorphous phases. The interphase regions in blends and co- polymers are important which depends on the chemical nature of the doping substances, the type and extent in which they interact with host matrix [10].

Since polymer blends are a mixture of chemically different polymers or copolymers so there are no covalent bondings between them [11]. In the recent past, the preparation of polymer blends is considerable to have importance in terms of the shorter time and lower cost of the product for the development than those of a new polymer [12]. The improvement of the performance of a polymeric material can be by selection of suitable elements and their ratios. Polymer blending produces certain new characteristics which lead to the formation of new materials with improved physical, mechanical and chemical properties [13, 14]. The new polymeric material has the properties of both the polymers. Polymer blends properties such as strength, toughness, etc tight relations with their micro phase morphology [15, 16]. A requirement for blends applications is not miscibility; it is an easy way to design a new polymeric material. Polystyrene (PS) is amorphous polymer; it is available with wide range of formations, which has radiation resistant and good thermal properties [17]. Poly methyl methacrylate (PMMA) is a transparent polymeric material possessing; it has many excellent properties such as high light transmittance, light weight, chemical resistance, good insulation, weathering corrosion resistance and colorlessness. PMMA is amorphous polymer which has good mechanical property. PMMA with PS have been form immiscible blend. Therefore, good mechanical properties can be achieved via blending of PMMA with PS [11].

The aim of this work is to study the nonlinear properties of olive oil films doped with PMMA, PS and their blend with different polymers concentration by using Z-scan technique. There are no detailed studies on the

nonlinear optical characterization of olive/polymer blending films. The work presented in this paper is an attempt to investigate in detail the nonlinear optical properties of the free casting olive oil/PS-PMMA films.

II. THEORY

2.1 Closed Aperture Z-Scan

Closed aperture Z-scan is technique for determining nonlinear refraction where the aperture is placed in the far field; the setup is shown in figure (1). In this technique, the sample is moving along the propagation direction (z) with fixed input energy. The normalized transmittance of the sample through the aperture in the far-field as a function of the position (z) is observed. The normalization is performed in a certain way so that when the sample far from focus, the nonlinearity is negligible and the transmittance is unity [18]. The intensity depends on the refractive index, which this relationship causes to change the beam radius of the transmitted beam but with retention of the Gaussian profile.

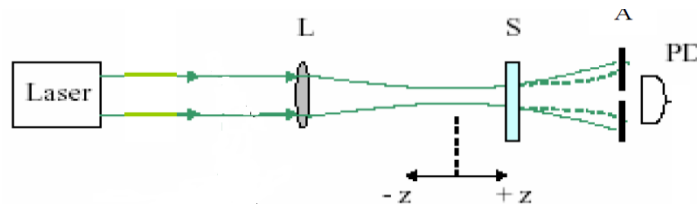


Figure (1): the scheme of closed aperture Z-scan.

In the Z-scan experiment, the transmittance variation can be given as follows: it can be start by scanning from a distance far away from the focus point (i.e., (-Z)), which the beam irradiance is low and negligible the nonlinear refraction leading to linear transmittance. As the sample is moving closed to the focus point, this cause to increase in the beam irradiance and produce self-lensing in the sample. In a self-defocusing (or negative self-lensing) before the focus point, the beam is collimating and the diffraction is reducing leading to a smaller beam at the aperture and the transmittance is increasing. As the scan continues which the sample crosses to (+Z) to the right of the focal plane, the same negative self-lensing effect will tend to increase diffraction and reduce the aperture transmittance. A peak (pre-focal transmittance maximum) and valley (post- focal transmittance minimum) will be, which the signature of Z-scan in negative and positive nonlinearity shown in figure (2). The negative one will give rise to an opposite valley-peak configuration than positive one [19, 20].

The change in transmittance between the peak and valley in the Z-scan can be defined as:

$$\Delta T_{pv} = T_p - T_v \dots (1)$$

where:

T_v and T_p : are the normalized valley and peaks transmittances.

The relation between the induced on axis phase shift and the change of transmittance between

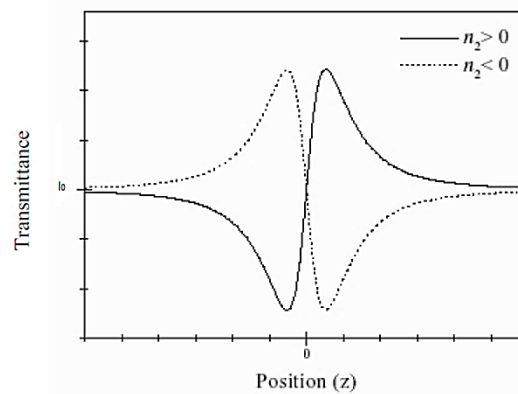


Figure (2): Calculated Z-Scan Transmittance Curves for a Third Order Nonlinearity, which the Positive Nonlinearity Shown by the Solid Line While the Negative Shown by the Dot Line.

the peak and valley for third order nonlinear refractive process in the absence of NLA (nonlinear absorption) is:

$$\Delta T_{pv} \cong 0.406 (1 - S)^{0.27} |\Delta\phi_0| \dots(2)$$

$$S = 1 - \exp(-2r^2 a/w_0^2) \dots(3)$$

where:

$\Delta\phi_0$: axis phase shift, S : is the aperture linear transmittance, w_0 : is the beam radius at the aperture in the linear regime [21].

It can be calculate the nonlinear refractive index from the peak to valley difference of the normalized transmittance by:

$$n_2 = \Delta\phi_0 / I_0 L_{eff} k \dots(4)$$

$$I_0 = 2P_{peak} / \pi w_0^2 \dots(5)$$

where:

I_0 : is on the axis irradiance, and L_{eff} : is the effective length of the sample, which can be determining from:

$$L_{eff} = (1 - e^{-\alpha_0 L}) / \alpha_0 \dots(6)$$

L : is the sample thickness, and α_0 : is the linear absorption coefficient, which can be determining from:

$$\alpha_0 = \frac{1}{L \ln(1/T)} \dots(7)$$

T : is linear transmittance [20].

The linear refractive index can be determined from:

$$n_0 = \frac{1}{T} + \left[\left(\frac{1}{T} \right) - 1 \right]^{0.5} \dots(8)$$

2.2 Open Aperture Z-Scan

The change in intensity of a beam focused by lens is measured by open aperture Z-scan, which the PD detector in the far field capture the entire beam and gives the value of the absorptive nonlinearity of a sample. The setup of open aperture Z-scan is shown in figure (3).

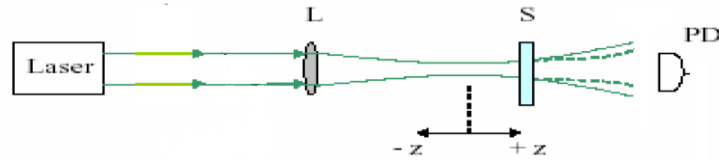


Figure (3): The Scheme of Open Aperture Z-Scan.

The absorptive nonlinearity can be caused either by:

1. Saturable absorption (SA), which at increase in the input laser intensity, the absorption coefficient decreases and as a result the transmittance increases; or
2. Reverse saturable absorption (RSA), which at increase in the input laser intensity, the absorption coefficient increases and as a result the transmittance decreases [22].

The normalized change in transmitted intensity can be given as [19]:

$$T(z) = \sum_{m=0}^{\infty} \frac{\left[\frac{\beta I_0 L_{eff}}{1 + \left(\frac{z}{Z_0}\right)^2} \right]^m}{(m+1)^{3/2}} \dots (9)$$

where:

$T(z)$: is the minimum transmittance, m : is integer, Z : is the sample position at the minimum transmittance, and Z_0 : is the Rayleigh range and it gives as:

$$Z_0 = n\pi w_0^2 / \lambda \dots (10)$$

The real and imaginary parts of the third order nonlinear optical susceptibility $[\chi^3]$ can be find from the value of the nonlinear refractive index and nonlinear absorption coefficient that determined experimentally according to the following [18]:

$$Re\chi^{(3)}(esu) = 10^{-4} \epsilon_0 c^2 n_0^2 n_2 / \pi \text{ (cm}^2/\text{W)} \dots (11)$$

$$Im\chi^{(3)}(esu) = 10^{-2} \epsilon_0 c^2 n_0^2 \lambda \beta / 4\pi^2 \text{ (cm}^2/\text{W)} \dots (12)$$

where:

n_2 : is the nonlinear refractive index, c : is the light velocity in vacuum, ϵ_0 : is the vacuum permittivity, and β : is the nonlinear absorption coefficient [18].

The absolute value of third order nonlinear optical susceptibility is given as:

$$|\chi^3| = [(Re(\chi^3))^2 + (Im(\chi^3))^2]^{1/2} \dots (13)$$

2.3 Self-Focusing and Defocusing

Kerr-Lasing or Self-Focusing is a result of the nonuniform spatial profile of the laser Gaussian beam. At high transmitted light intensity, the change of refractive index will modify the light propagation not only with relation to the polarization but in its geometrical properties also [23].

The Kerr-Lens focal length for the Gaussian beam of radius ω_0 is given as:

$$F = a\omega_0^2 / 4Ln_2I \dots (14)$$

Where:

a : is the correction term, I : is the irradiance, and L : is the thickness of the nonlinear (sample).

When the nonlinear refractive index n_2 is negative, then the focal length according to the above equation will be negative and thus the incident beam is the self-focusing [19].

2.4 Optical Limiting

In a material that has a strong nonlinear effect, as the input intensity increase then the absorption of light increase. At which exceed a certain input intensity, the output intensity accesses a constant value. Like this material can be used to limit the amount of optical power that entered to the system. The ideal behavior of such system is shown in figure (4) [24]. The important utility in evaluating of optical limiting material is whether it exhibits broad band spectral response, i.e. at low intensities; the material is transparent while at high intensities it is behaving a large nonlinearity over a broad band spectral range.

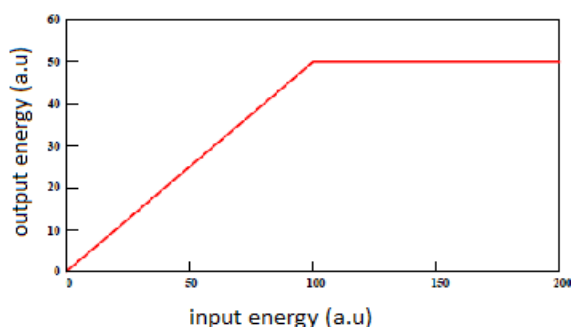


Figure (4): An ideal optical limiter [25].

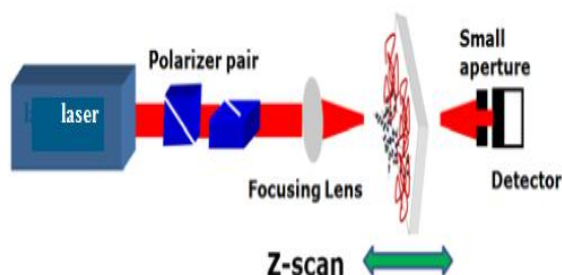


Figure (5): The scheme of optical limiting [26].

III. EXPERIMENTAL SECTION

Olive oil is an organic material of complex compound made of glycerides, fatty acids, sterols, wax esters, uvaol, erythrooil, phenolic compounds, tocopherols, aroma components, aromatic hydrocarbons, tocopherols, xenobiotics, water soluble components, unsaponifiable matter and microscopic bits of olive [27].

Olive oil is considered an organic compound because of its two π electron bound, or two double bound called conjugated or higher so material in this condition called dye material. Olive oil contained many organic compounds. Two of its lye under dye material condition where these two compounds are:

a) Linoleic polyunsaturated (two double bound) ($C_{17}H_{29}COOH$) or $CH_3-(CH_2)_4-CH=CH-CH_2-CH=CH-(CH_2)_7-COOH$ Figure (6). [27, 28]

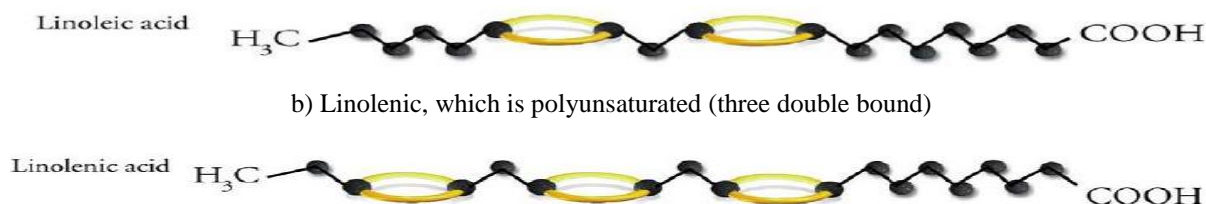


Figure (6): organic compounds of Olive oil.

Two types of polymers had been used in this study: Poly Methyl Metha Acrylate (PMMA) and Polystyrene (PS). They are the transparent plastic material with high elasticity, index of refraction. Their resistance to climate changes is better than other polymers, and their optical properties are very good [29].



Polymethylmethacrylate (PMMA) and Polystyrene (PS) are from ICI Company, were reported to have molecular weights of 145,000 and 167,000 $\text{g} \cdot \text{mol}^{-1}$, respectively. Tetrahydrofuran (THF): from (BDH) laboratory reagents (BDH Chemicals Ltd Poole England).

The polymer PMMA ((C₅O₂H₈)_n) is a thermal plastic polymer which means that if it is heated to a degree not exceeding its melting point and then cooled to its previous temperature, it will keep most of its properties especially its transparency (92%) and its amorphous molecular structure[29].

IV. SAMPLE PREPARATION

Each volume of pure olive oil was dissolved with the same volume of Ethanol and was stirring for half hour to get homogenous solution.

For the preparation of olive oil films doped with PMMA ,PS and their blend :1gm of PMMA ,1 gm of PS was dissolved in 10ml of THF separately to obtain a 10% w/v solution after one hour of stirring.

A homogenous solution of olive oil and ethanol was added to the two homopolymers and also their blend (PS/PMMA) with different ratios of polymers. The ratio of doping olive oil to PMMA, PS and their blend are:1/2 (50%), 1/3 (33 %), 1/4 (25%).The prepared solutions were stirring to mix very well.

Doped polymer films were fabricated by the free casting technique (FC), which this technique includes casting a polymer solution in petri dish (flat-bottomed glass cup) without imposed the hydrodynamic stress on liquid. In order the dishes have a plane situation, there have been arranged on a glass plate, and then it covered the dishes by a heavy paper box in order to protect it from light and dust. Under ambient condition (30 °c), the solvent is allowed to evaporate until the films harders. Then the harder films were removed from the petri dish by first washing it off with distilled water and then placed it in the oven at (50 °c) temperature for 10 minutes to dry.

The thicknesses of the films were measured with an electrical device (Mini-test 3000 microprocessor coating thickness) from electro, physik, Germany (ERICHSEN).The films had thickness of (0.00135-0.0035) cm. The experimental work based on the testing of olive oil doped films absorption spectrum using UV-VIS spectrophotometer. The Z-scan experiments were performed using a 650 nm CW diode laser source. Maximum power is 100 mW, beam divergent 0.045 mrad, Ac: 220-240 volt, frequency: 50-60 Hz 250 mA that focused by 10 cm focal length lens. The laser beam waist ω_0 at the focus is measured to be 0.00454 mm, and the Rayleigh length to be 0.9957 cm.

V. RESULTS AND DISCUSSION

UV-Visible absorption spectra of pure olive oil and olive oil doped films with PMMA, PS and their blend with different ratio of olive oil to polymers (1/2 (50%),1/3 (33 %), 1/4 (25%)).Were carried out by (Shimadzu UV-1601PC spectrophotometer). Which operates in wavelength range of 200-1100 nm .

Figs.7 show the absorption spectrum at room temperature for pure olive oil. Fig. (8-10) show the absorption spectra of olive oil doped films with PMMA, PS and their blend with different ratio of olive oil to polymers (1/2 (50%),1/3 (33 %), 1/4(25%)) respectively. The films are found to be highly transparent throughout the visible region compared with pour olive oil. Furthermore, the absorption peak wavelengths are substantially blue shifted relative to that of pure olive oil due to the strong confinement effect [30]. The presence of olive oil, thus, enhances the UV absorption of the composite films and modifies the overall optical behavior of polymer blend

films [31]. The absorption window is found in the range 625-700 nm. Also, it can be seen that the absorption peak value for olive oil doped films increased as the ratio of olive oil increased due to the increase in the number of the absorbing species according to Beer's law [31, 32]. The large absorbance of different samples at 650 nm confirms that nonlinear effects may be observed at the applied wavelength.

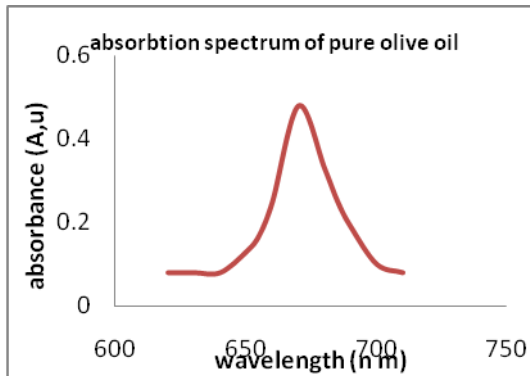


Figure 7: Absorption spectrum of pure Olive Oil films

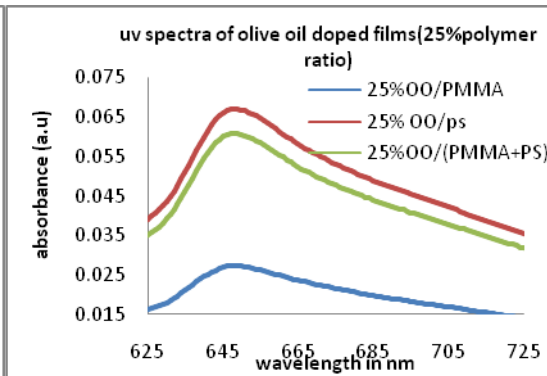


Figure 8: Absorption spectra of 25% Olive oil doped films

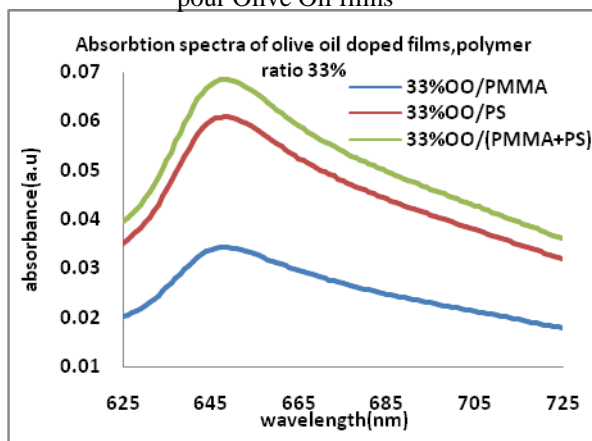


Figure 9: Absorption spectra of 33% Olive oil doped films

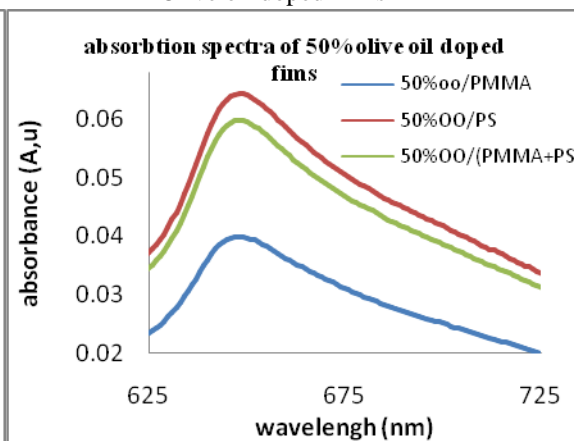


Figure 10: Absorption spectra of 50% Olive oil doped films

Nonlinear optical properties (the third-order nonlinear refractive index n_2 and the nonlinear absorption coefficient β , third order susceptibility(χ^3) of olive oil doped films with PMMA, PS and their blend were investigated by Z-scan technique (open and closed aperture) by using cw diode laser with 650 nm and incident intensity $I_0 = 1545.107 \text{ watt/cm}^2$.

In the case of open aperture Z-scan when the aperture is removed any nonlinear absorption present in the sample can be found in the measurement. In this case, the sample transmittance is measured as a function of intensity when the sample is scanned through the laser beam focal plane. If the nonlinear absorption coefficient β is positive, it increases with increasing the input power irradiance (two photons absorption), but if β is negative, it decreases with increasing the input power irradiance (saturation absorption)[33].

Fig. 11 (a, b and c) :shows open aperture Z-scan curves for the different ratio of olive oil (OO) (25%, 33%, and 50%) doped with PS, PMMA and their blend. The Z-scan curve shows negative signs for the absorptive nonlinearities, upward peak indicative of the saturation of absorption. The transmittance of the sample increases as it brought close to the focus of the z- scan lens. This is in sharp contrast to the recent studies on pure olive oil [34], where the corresponding curve is a valley indicating that nonlinear absorption increases with intensity and transmission falls with intensity. Saturable absorption observed, in this case, is similar to the behavior of yellow

disperse doped PMMA [35] and a Complex of Plasmonic and Molecular-Like Au Nanocrystals [36], under similar experimental conditions.

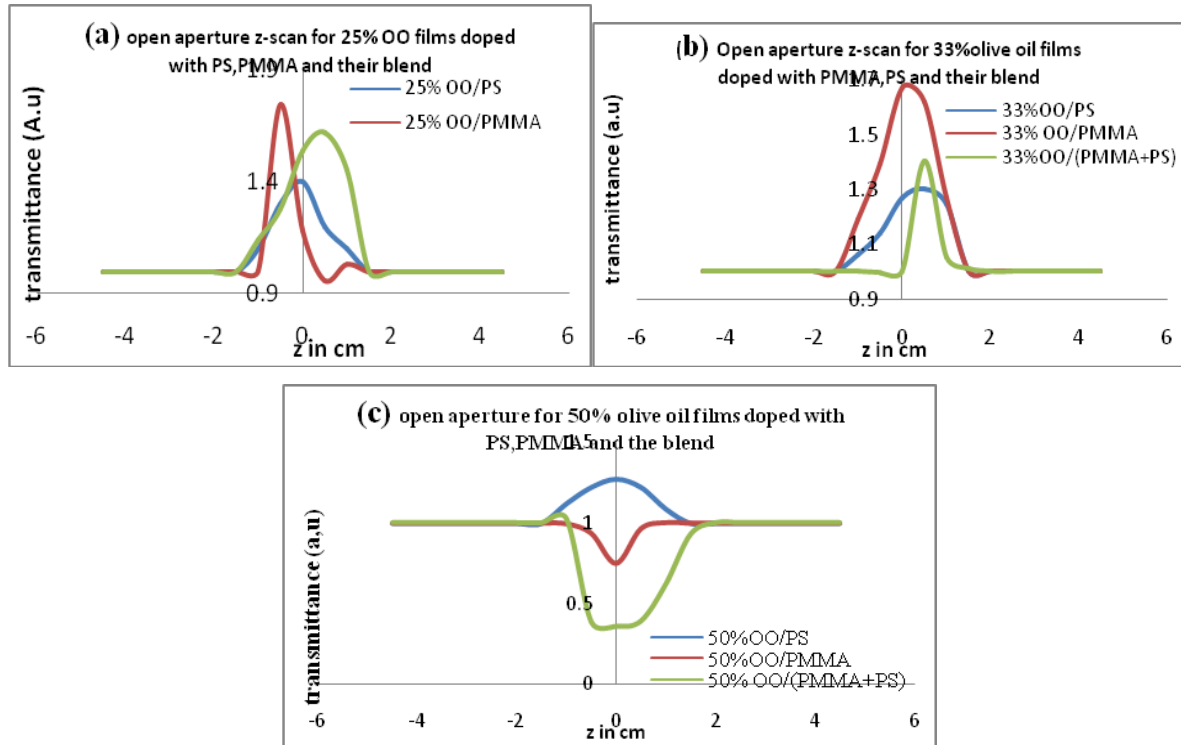


Fig.11: Open aperture z-scan of different ratio of Olive oil (OO) films doped with PS,PMMA,(PMMA+PS).(a):25%,(b):33% and 50%

From open aperture Z-scan curve, it is found that films of 25%, 33%, exhibited saturation absorption behavior. Films of 50% OO/PS still has saturation absorption behavior but the nonlinear behavior of 50% OO/PMMA and 50% OO/ (PMMA+PS) alters from the saturable absorption (SA) to reverse saturable absorption (RSA) as the ratio of doping is increased. The transformation from SA to RSA suggests that another nonlinear process takes place and becomes dominant which could be probably due to either two-photon absorption (TPA) or excited state absorption (ESA). Such an interesting effect can be used for optical pulse compressor, optical switching, and laser pulse narrowing [37]. Such kind of behavior has been reported earlier for different molecules including Rhodamine B [38], solid Poly-methyl-methacrylate (PMMA) composite containing porphyrin-covalently [39], nickel sulfide nanoparticles [33], they argue the presence of such behavior could be due to: most of the particles occupy the excited state. As the ground state is bleached [29], the system becomes increasingly transparent to the incident laser pulse resulting in a saturation of absorption and the consequent Z-scan curve. From the open aperture Z-scan curve, it is found that the blending of PMMA with PS moderate the films transmittance occurs near the focal position compared to OO/PS and OO/PMMA.

Table (1): Nonlinear parameters of open aperture z-scan for olive oil (OO) doped films with PMMA, PS, and their blends with different ratio of olive oil by using CW diode laser at 650 nm

Films	25%			33%			50%		
	T	β w/cm	$Im(x^3)$ w/cm	T	β w/cm	$Im(x^3)$ w/cm	T	β w/cm	$Im(x^3)$ w/cm
OO/PMMA	1.74	2.22	5.96	1.67	1.88	5.49	1.27	0.76	2.36
OO/PS	1.4	2.89	11.17	1.3	2.46	9.52	0.75	0.71	2.48
OO/(PMMA+PS)	1.62	1.67	6.44	1.4	1.27	5.27	0.36	0.28	1.09

From table (1) we noticed that: the value of β and $Im(x^3)$ for all doped films with a different ratio of doping decreased as the concentration of polymer increased. Doping materials generally exhibit a range of properties varying between the properties of their components. Moreover, their properties may be complementary and difficult to be found together in the case of a single component [40, 41]. This mean that the doping and blending of two polymers do not improve the nonlinear optical properties and that because olive oil contained many organic compounds.

Fig. 12 (a, b and c) :shows closed aperture Z-scan curves for the different ratio of olive oil (OO) (25%, 33%, and 50%) doped with PS, PMMA their blend by using CW diode laser ay 650 nm. The closed-aperture curve exhibits a valley to peak shape, indicating a positive value of the nonlinear refractive index n_2 that which results from self-focusing that confirms the nonlinear absorption behavior [5].

This is in sharp contrast to the recent studies on pour olive oil [34], where the corresponding curve exhibits a peak to valley shape, indicating a negative value of the nonlinear refractive index n_2 . Such kind of behavior has been reported for different molecules including Ternary Oxides barium strontium titanate $BaxSr_{1-x}TiO_3$ _BST_ thin films [41] and ZnSe Quantum Dots in Glass-Matrix Thin Films [43].

Absorption saturation and positive refractive index explained by [19]: Consider a homogeneously broadened system of two-level atoms, Figure (13). The refractive index is positive for frequencies below resonance and negative at frequencies above it, assuming that the rate at which photons are supplied to the system produces a negligible change of population in the upper and lower states. This means that the rate of population relaxation (recombination and diffusion, for example) is much faster than excitation. When the incident irradiance is sufficiently high, however, this may no longer be the case. The upper level can become appreciably occupied, reducing the availability of terminal states for optical transitions. The absorption thus decreases or bleaches, indicated by the dashed lines in Figure (12). Associated with the change of absorption is a change of refractive index.

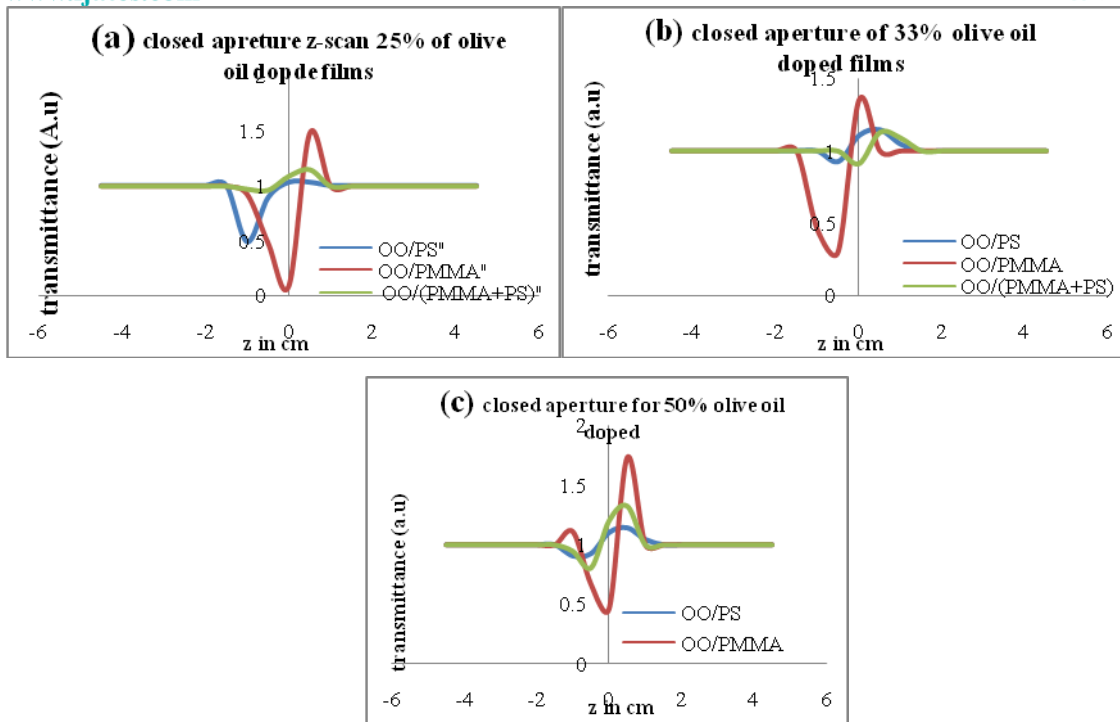


Fig.12: Closed Aperture z-Scan of Different Ratio of Olive oil (OO) Films Doped with PS,PMMA,(PMMA+PS).(a):25%,(b):33% and 50%

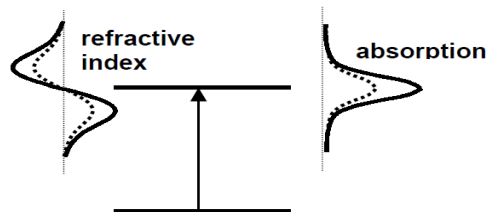


Fig.13:A System of Two Level Atom to Explain Saturation Absorption and Positive Refractive Index(self focusing)[19]

From table (2) we noticed that: the value of n_2 , $R(x^3)$ and χ_3 for all doped films with different ratio of doping decreased as the concentration of polymer increased and the enhancement of the refractive index was believed to be due to the increased thermal effect with CW pumping. The energy from the focused laser beam is transferred to sample through linear absorption and is manifested in terms of heating the medium leading to a temperature gradient and there by the refractive index change across the sample which then acts as a lens. The phase of the propagating beam will be distorted due to the presence of this thermal lens. The peak–valley separation of more than 1.7 times the Rayleigh range of 0.475 mm in our experiment also suggests the presence of the thermal component. It is well established that a separation of $\sim 1.7z_0$ indicates Kerr-type of nonlinearity [26, 44].

Table (2): Nonlinear parameters of Ox./PMMA, Ox./PS ,Ox./PMMA-PS films by using CW diode laser at 650 nm for open aperture z-scan

Films	25%			33%			50%		
	$n_2 \times 10^{-5} \text{ cm}^2/\text{W}$	$R(x3) \times 10^{-4} \text{ cm}^2/\text{W}$	$\chi^3 \text{ cm}^2/\text{W}$	$n_2 \times 10^{-5} \text{ cm}^2/\text{W}$	$R(x3) \times 10^{-4} \text{ cm}^2/\text{W}$	$\chi^3 \text{ cm}^2/\text{W}$	$n_2 \times 10^{-5} \text{ cm}^2/\text{W}$	$R(x3) \times 10^{-4} \text{ cm}^2/\text{W}$	$\chi^3 \text{ cm}^2/\text{W}$
OO/PMMA	11.2	65.1	5.96	8.16	37.7	5.49	12.9	100	2.36
OO/PS	2.68	5.38	11.17	1.24	1.15	9.52	1.25	1.05	2.48
OO/(PMMA+PS)	1.856	2.58	6.44	5.89	27.8	5.27	6.56	31.8	1.09

The limiting effect of olive oil doped films was studied by using a 100 mW, CW laser at 650 nm. The experimental set-up for the demonstration of optical limiting is shown in Fig. 4. OL can be achieved through various nonlinear optical mechanisms such as multi-photon absorption (MPA), excited state absorption (ESA), free carrier absorption (FCA), self-focusing, self-defocusing, nonlinear scattering, photo-refraction etc [44,45].

Olive oil doped films were kept at the position where the transmitted intensity shows a valley in closed aperture Z-scan curve. A polarizer was used to vary the input power. The input laser intensity is varied systematically, and the corresponding output intensity values were measured by the photodetector. At very high peak intensities (closer to the focus) we could observe diffraction type pattern with concentric ring structures probably due to self-phase modulation. However, in limiting experiments we have ensured that there is no ring pattern formation by placing the sample away from focus.

Figure (14a, b, and c) show the optical limiting for olive oil doped films at different concentration, where the films of OO/PMMA does not exhibit any optical limiting for the three doping ratio, but films of OO/PS and OO/(PMMA+PS) with the different ratio of doping have very good optical limiting behavior. The output power rises initially with increasing input power, but after a certain threshold value the samples start defocusing the beam, resulting in a greater part of the beam cross-section being cut off by the aperture. Thus, the transmittance recorded by the photo detector remained reasonably constant showing a plateau region and is saturated at a point defined as the limiting amplitude. i.e., the maximum output intensity, showing obvious limiting property.

For the three doping ratio the threshold power value of optical limiting is approximately about 45 mW with little change in the output intensity as the polymer concentration is changed special films of blending polymer which mean blend polymer can improved the optical limiting effect, table (3) . Since the samples are pumped with CW laser beam the arising nonlinearities are predominantly thermal in nature because its nonlinear absorption coefficient increases with an increase in the incident irradiance.

Table (3): Optical Limiting of Olive Oil Polymer Doped Films (Output Limiting Amplitude and Input Threshold)

films	Input threshold	Output Limiting amplitude	Input threshold	Output Limiting amplitude	Input threshold	Output Limiting amplitude
	25%		33%		50%	
OO/PS	40	9.1	40	9.2	45	9.9
OO/(PMMA+PS)	45	8.1	45	12.2	45	12.3

Since the thermal expansion is large, high absorbance of the nonlinear material at the corresponding wavelength leads to an increase in temperature and density of the sample. Heating due to laser absorption is responsible for changing the absorption coefficient and optical limiting effect.

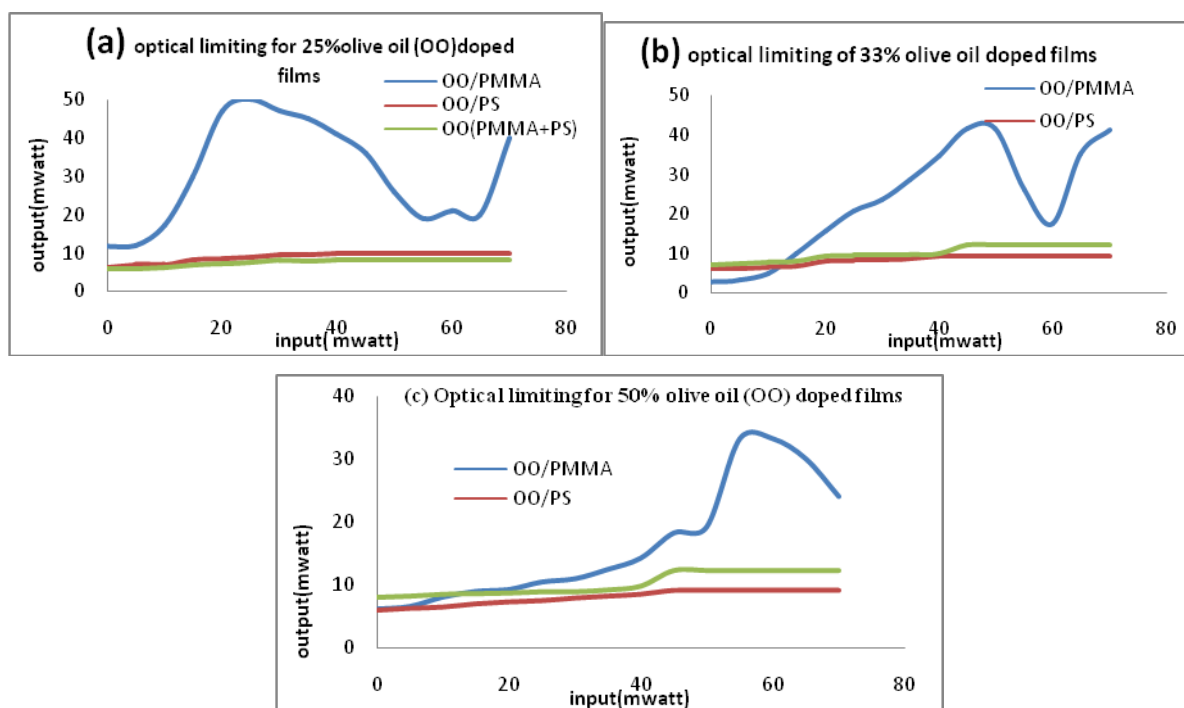


Figure (14): Optical Limiting Effects of Olive Oil Doped Films for Closed Aperture S-Scan, (a) OO, 25%, (b) OO 33%,(c) OO 50%

VI. CONCLUSION

The nonlinear properties (absorption, refraction and optical limiting) for olive oil doped film with PMMA, PS and their blend with different ratio of concentration (25%, 33%, and 50%) were studied using a single-beam Z-scan technique under CW laser at 650 nm. It is noting that the value of χ^3 for the doped films is larger than pour olive oil and those of some representative third-order nonlinear optical materials indicating promising prospects for nonlinear optical applications. From open aperture Z-scan curve it is found most the films exhibited saturation absorption behavior except films of 50% OO/PMMA and 50% OO/ (PMMA+PS) alters from the saturable absorption (SA) to reverse saturable absorption (RSA) as the ratio of olive oil is increased. This is in

sharp contrast to the recent studies on pour olive oil , where the corresponding curve exhibits reverse saturation absorption, indicating a positive value of the nonlinear absorption coefficient β_2 (two-photon absorption). From closed aperture Z-scan curves all the films with the different ratio of olive oil exhibits a valley to peak shape, indicating a positive value of the nonlinear refractive index n_2 which results from self-focusing that confirms the nonlinear absorption behavior. This is also in sharp contrast to the recent studies on pure olive oil, where the corresponding curve exhibits a peak to valley shape, indicating a negative value of the nonlinear refractive index n_2 and films nonlinearity arising from thermal origin where laser absorption is responsible for changing the absorption coefficient and optical limiting effect. The nonlinear parameters (n_2 , β_2 , IM (χ^3), $R(\chi^3)$ and χ^3) of olive oil doped films with different concentration of polymers with films of blending polymer decreased as the concentration of polymer increased due to olive oil aliphatic chain and active hydroxyl group .

But these films exhibit very good optical limiting properties, which mean blend polymer can improved the optical limiting effect.

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