RESEARCH PAPER

Nonlinear Optical Properties of Mixed Organic Laser Dye Thin Films Doped with Metal Nanoparticles and PMMA Polymer

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ABSTRACT

This work investigates the non-linear optical properties of thin films composed of an organic laser three-dye combination (Fluorescein, Eosin-Y and Rhodamine B) doped with PMMA polymer and incorporating (Ag, Cu, and SiO_2) nanoparticles dissolved in chloroform solvent at room temperature, The thin films were prepared by drop-casting dye solutions at a concentration of 10^{-3} M. The absorption spectra were recorded in the (300-800) nm range. X-ray analysis was utilized to determine the structure and composition of materials. Where the(Mix+ PMMApolymer +Ag nanoparticles) exhibited the highest crystallinity. The AFM assay results for thin films after deposition of the polymer-based dye mixture and nanomaterial's revealed that as the film thickness increased, the surface roughness and average diameter increased, thus increasing the linear absorbance, which in turn increased the nonlinear absorption coefficient. The nonlinear optical parameters, including the nonlinear absorption coefficient (β) and nonlinear refractive index (n_2), were measured using the open- and closed-aperture Z-scan technique. The open-aperture results confirmed that the saturable absorption was present in all thin films. Strong nonlinear optical responses were observed, with a linear dependence of the nonlinear refractive index on the nonlinear phase shift. The results suggest that the prepared films are suitable for use as an active laser medium and in optoelectronic application.

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INTRODUCTION

Nonlinear optical (NLO) materials have attracted intense scientific interest due to their critical significance in modern photonic and optoelectronic applications such as optical switching, optical modulation, optical limiting, and ultrafast data processing. Organic dyes integrated into polymer matrices have emerged as viable candidates for NLO materials due to their large nonlinear optical coefficients, rapid reaction times, mechanical flexibility, and simplicity of thin-film fabrication [1].

Fluorescein, Eosin-Y and Rhodamine B dyes are well-known organic chromophores that exhibit great visual absorption, high fluorescence efficiency, and significant light-matter interaction. Combining these dyes can improve spectral coverage and nonlinear optical sensitivity via energy transfer mechanisms and cooperative interactions

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between dye molecules [2]. When dissolved in an appropriate solvent, such as chloroform, and embedded in a transparent polymer matrix, these dyes can generate optically uniform and stable thin films suited for nonlinear optical experiment. Poly(methyl methacrylate) (PMMA) is widely used as a host polymer because of its high optical transparency, thermal and chemical stability and compatibility with a wide range of organic and inorganic dopants. Incorporating dye mixture into PMMA offers structural stability and avoids dye aggregation, therefore improving the optical quality and repeatability of the prepared thin film [3]. In recent years, it has been demonstrated that incorporating metallic nanoparticles, such as silver (Ag) and copper (Cu) nanoparticles, into dye-polymer composites greatly improves nonlinear optical characteristics. This boost is mostly due to localized surface plasmon resonance (LSPR), which causes significant local electromagnetic field amplification at the nanoparticle surface. The interaction between plasmonic nanoparticles with dye molecules can cause higher nonlinear absorption and refractive index shifts, boosting the efficiency of nonlinear optical processes [4]. The phenomenon of surface Plasmon resonance (SPR) explains Nonlinear optical effects and selective optical absorption within the same spectral range, from the collective oscillation of free electrons at the surface of metallic nanoparticles upon excitation by incident light.

An optical limiter is a nonlinear optical device or material designed to protect sensitive optical components and the human eye from damage caused by high-intensity light sources, such as powerful laser radiation. It allows low-intensity light to pass through with minimal attenuation, while significantly reducing transmission of light when the incident intensity exceeds a threshold [5,6]. The aim of this research is studying the effect of mixing of three organic laser dyes to obtain a new dye and its deposition thin films using (PMMA) polymers with added (Ag, Cu and SiO_2) nanoparticles and Possibility of using this mixture organic laser dyes as optical limiting and active laser medium.

THEORY

In this work, thin films were prepared by dissolving a combination of (Fl, E-Y, and RB) dyes in chloroform and embedding them in a PMMA matrix containing silver and copper nanoparticles. The nonlinear optical characteristics of the produced thin films were thoroughly examined to determine their suitability for nonlinear optical and photonic applications. The findings provide important information on the synergistic effects of dye mixtures, polymer hosts, and metallic nanoparticles on the nonlinear optical behavior of composite thin films.

The refractive index (n) and optical absorption (α) may be found as functions of the incident laser beam's intensity (I) using Maxwell's equations as Eq. 1 [7]:

$$\alpha = \alpha_o + \beta I \tag{1}$$

where I represents the incoming intensity, α_o is the linear absorption coefficient, and β is the intensity-related nonlinear absorption coefficient. To find the linear refractive index at high intensity the following equation is utilized (Eq. 2) [8]:

$$n = n_o + n_2 I \tag{2}$$

where n_0 is the linear refractive coefficient, and n_2 is the nonlinear refractive index. To examine the nonlinear optical characteristics, the Z-scan technique determines the nonlinear refractive index in a closed-aperture configuration using the Eq. 3 [9]:

$$n_2 = (\Delta\phi_\circ)/(I_0 l effk) \tag{3}$$

where $\Delta\Phi_o$ is the nonlinear phase shift given by Eq. 4 [10]:

$$\Delta T_{p-v} = 0.406|\Delta\Phi_0| \tag{4}$$

where $\Delta Tp{-}v$ is the normalised transmittance difference, which can be computed using a closed-aperture Z-scan setup, between the transmittance values at the top and the valley, k = 2π/λ, where (λ) denotes the spectrum of the beam, (I_o) represents the intensity level at the focal point (Eq. 5) [9].

$$I_o = \frac{2P_{peak}}{\pi \omega_\circ^2} \tag{5}$$

Where (ω_o) is the beam radius at the focal point equal 0.025 mm.

The Eq. 6 can be used to compute the nonlinear absorption coefficient

$$\beta = \frac{2\sqrt{2}\ T(z)}{I_o L_{eff}} \tag{6}$$

Where T(z) is the lowest normalized transmittance at the focal point. To determine the effective length (L_{eff}) of an optical medium at (Z=0), the Eq. 7 is used [11]:

$$L_{\text{eff}} = \frac{(1 - \exp^{-\alpha_0 L})}{\alpha_0} \quad (7)$$

where L is the length of a sample and α_0 is the linear absorption coefficient.

MATERIALS AND METHODS

Fl, E-Y, and RB are among the most visible xanthene dyes [12]. These organic dyes exhibit strong fluorescence and photostability. These xanthene family members have gained popularity due to their self-association behavior in solutions and other applications. They are commonly employed in biomedical imaging, histological staining, fluorescence microscopy, and laser systems. The molecular weight of E-Y ($C_{20}H_6Br_4Na_2O_5$) is 691.9 g/ml, Fl ($C_{20}H_{12}O_5$) is 332.31 g/ml and RB ($C_{28}H_{31}N_2O_3Cl$) is 479.02 g/ml. The dyes were purchased from Sigma-Aldrich with purities of more than 99%. [13]. Fig. 1 depicts the molecular structure of the dyes.

Chloroform was used as a solvent. It is largely unreactive and miscible with most organic liquids. The molecular structure of chloroform solvent is $CHCl_3$, with a molar mass of 46.07 g/ml and a polarity of 0.5771 [14]. PMMA (polymethyl methacrylate) polymer is known as acrylic or acrylic glass, and it also goes by the commercial name Plexiglas. The molecular structure of PMMA polymer is $CH_2{=}C(CH_3)COOR$, with a molecular weight of 84000 g/ml [15].

Silver nanoparticles range in size from 1 to 100 nanometers. Despite being referred to as “silver”, some include a significant amount of silver oxide due to their high surface-to-bulk ratio. The form of the nanoparticles may differ depending on the purpose of the inquiry. Sigma-Aldrich manufactures Nano powders with a surface area more than 40 m^2/g [16]. The copper nanoparticles were supplied by Laboratory Reagent, Ltd. Copper nanoparticles are highly sought after due to their superior thermal

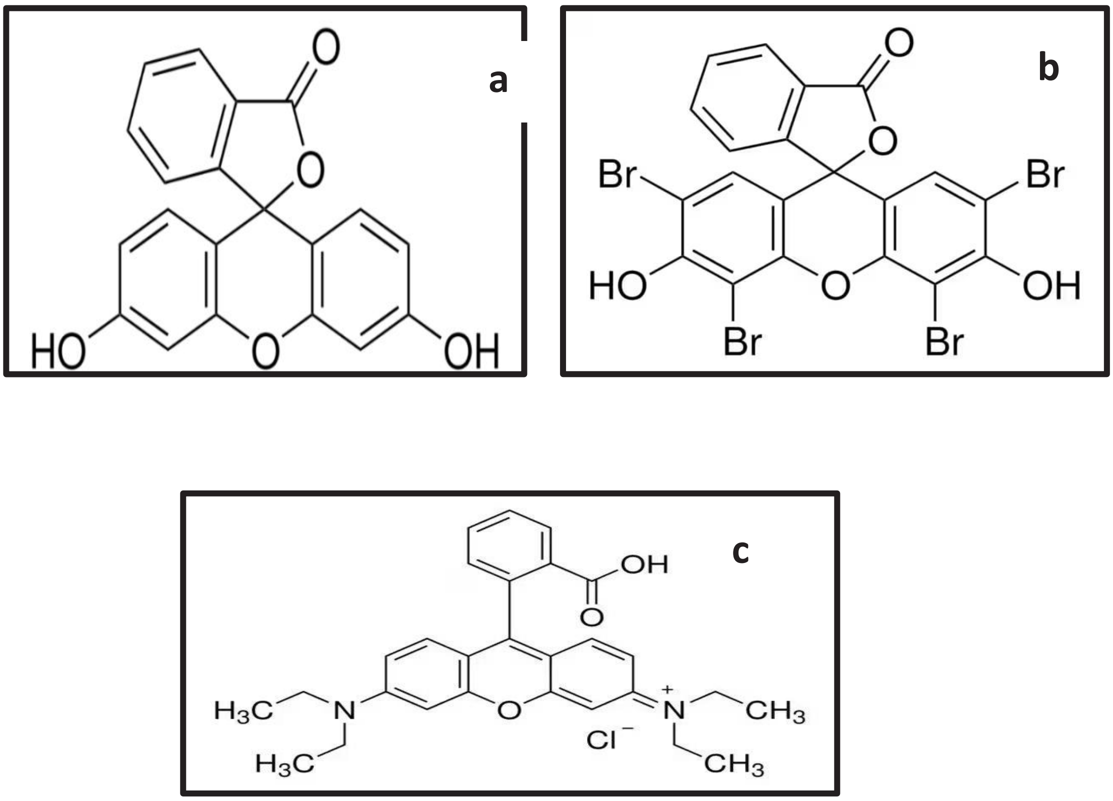

Fig. 1. (a)The Molecular Structure of Fluorescein Dye (b)The Molecular Structure of Eosin Dye (c) The Molecular Structure of Rhodamine B dye [13].

conductivity and other heat transfer properties. Copper nanoparticles also have larger surface-area-to-volume ratios, lower manufacturing costs, antibacterial efficacy, catalytic activity, and optical and magnetic properties. The primary issue lies in their preparation and preservation, as they readily oxidize upon exposure to air. The circumference ranged from 40 to 70 m^2/g, while the layer thickness was between 10 and 100 nm [17].

Silicon dioxide (SiO_2) nanoparticles are commonly utilized nanomaterials with particle sizes ranging from 1 to 100 nm. SiO_2 nanoparticles, with their high surface-to-volume ratio, have distinct physical and chemical characteristics compared to their bulk counterpart. They are very thermally stable, chemically inert, optically transparent, and dispersible in a variety of solvents and polymer matrices. The properties of SiO_2 nanoparticles make them suitable for use in optics, photonics, coatings, and nonlinear optical materials. SiO_2 nanoparticles can boost light transmission, minimize scattering losses, and improve optical uniformity in nanocomposite thin films [18].

Fl, E-Y, and RB were combined with PMMA polymer, silver nanoparticles (AgNPs), copper nanoparticles (CuNPs), and silicon dioxide nanoparticles (SiO_2NPs) to create thin films on a clean glass slide using the drop-casting method. The films were dried at room temperature (25–30°C) for two days after each solution was produced at a concentration of 10^{-3} M. The thickness of these thin films varies between 110 and 200 nm. Employing an optical method and an optical thin film measurement model, the thin-film thickness was measured by LIMF-10. A magnetic stirrer was used to mix the polymer solution with the appropriate amount of dye solution after the necessary amount of polymer (2g in 30ml of chloroform solvent) was dissolved to create a homogenous mixture. To create a uniform mixture, 0.02g of silver (Ag), copper (Cu), and

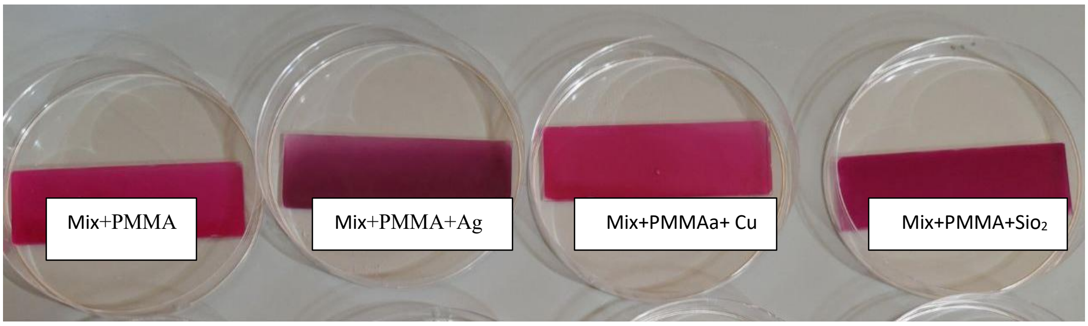

Fig 2. Thin films of Fl, E-Y, and RB mixture with PMMA polymer incorporating silver, copper, and Silicon dioxide nanoparticles.

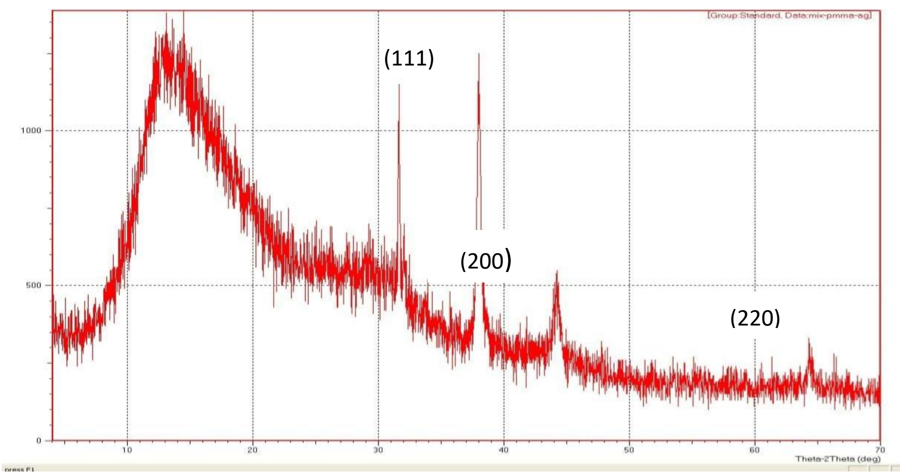

Fig. 3. XRD patterns of Mix+PMMA+AgNPs, films.

(SiO_2) nano powders were added and combined using a magnetic stirrer, as shown in Fig. 2.

RESULTS AND DISCUSSION

An XRD instrument was used to explore the various structural properties of thin film samples made from a mixture of organic laser dyes doped with polymer and nanoparticle. The XRD peaks were found between 2θ=20° and 2θ=70°. According to the International Center of Diffraction Data (ICDD) card number 26-0575.

The X-ray diffraction (XRD) patterns of the prepared samples are shown in the figures (3,4 and 5.) The polymer/dye film exhibits a broad diffraction band around 2θ ≈ 18–22°, which indicates the amorphous nature of the polymer matrix. After incorporating the nanomaterial's, additional diffraction peaks appear in the patterns, confirming the formation of crystalline phases within the polymer matrix.

In the Fig. 3. shows distinctive peaks at 2θ = 38.1°, 44.3°, 64.4°, and 77.5°, which correspond to the (111), (200), (220), and (311) crystallographic planes of face-centered cubic (FCC) silver nanoparticles [19]. Meanwhile, Fig. 4. displays copper nanoparticles with new diffraction peaks at 2θ = 43°, 50°, and 74°. These peaks correspond to the (111) and(200) crystallographic planes of copper, suggesting the creation of crystalline Cu nanoparticles inside the polymer matrix [20].

Fig. 5. A diffraction peak observed at 2θ ≈ 31.6° (d ≈ 2.82 Å) is indexed to the (102) plane

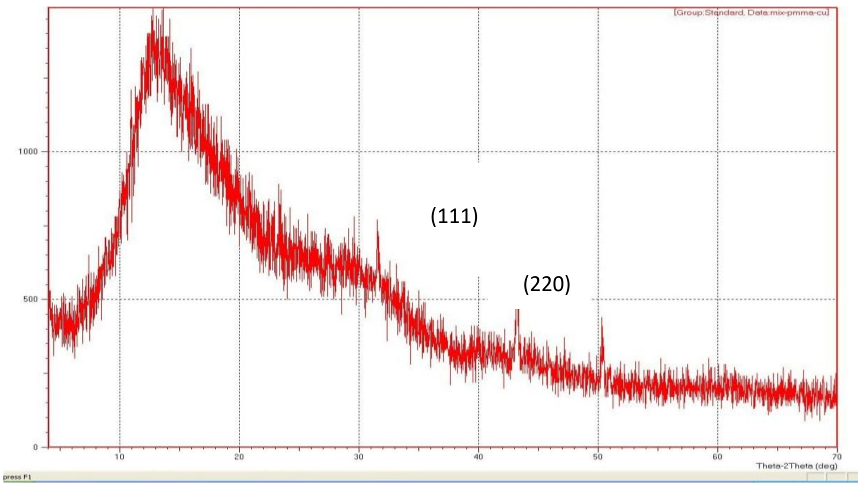

Fig. 4. XRD patterns of Mix+PMMA+CuNPs, films.

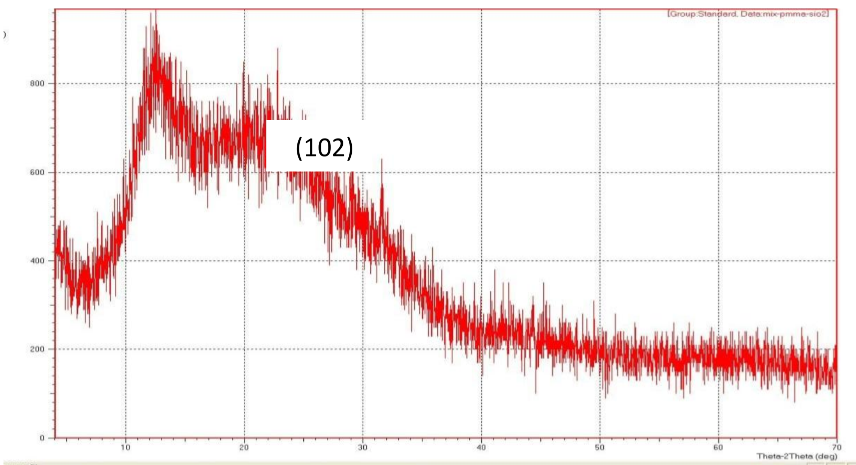

Fig. 5. XRD patterns of Mix+PMMA+SiO_2NPs, films.

of SiO_2. The relatively low intensity and peak broadening are attributed to the dispersion of SiO_2 nanoparticles within the amorphous PMMA matrix and the presence of organic dyes.

In Table 1. the XRD data reveal that the crystallite size of the films reduces following the inclusion of metal nanoparticles such as silver and copper into the polymethyl methacrylate (PMMA) matrix. Consequently, smaller crystallite sizes lead to a larger surface area and stronger interaction between the material and the incident light. This behavior enhances the nonlinear optical response of the nanocomposite films, making them suitable for various optical applications such as optical limiting and photonic devices[21]. It was calculated from the full width at half maximum (FWHM) (β) of preferential orientation diffraction peak by using the Debye-Sherrer̓ s equation (Eq. 8).

$$D = \frac{0.94\ \lambda}{\beta \cos\theta} \quad (8)$$

Where: D: is crystallite size, β: is (FWHM) in radians

Fig. 6. shows 3D AFM images of thin films prepared from the mixture of three organic laser dye (Fl, E-Y and RB) doped with PMMA polymer and AgNPs, CuNPs and SiO_2NPs at 10^{-3} M. Table 2. shows the average particle sizes in these thin films. It is noticed that the root mean square

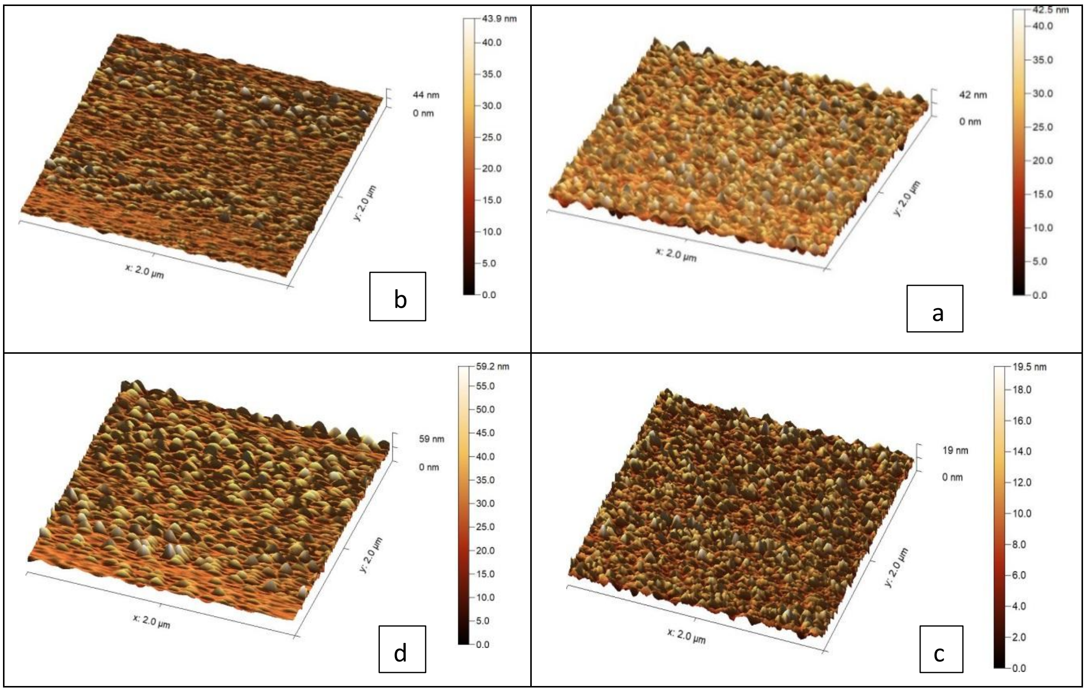

Fig. 6. 3D AFM images of thin films generated from a mixture of organic laser dyes doped with polymer and nanoparticles (a). Mix+PMMA, (b) Mix+PMMA+SiO_2NPs, (c) Mix+PMMA+CuNPs, (d) Mix+PMMA+AgNPs.

Table 1. XRD parameters of a combination of PMMA polymer thin films doped with Ag, Cu and SiO_2 nanoparticles.

Sample	(hkl)	2Θ (Deg.)	Θ (Deg.)	FWHM (rad)	D (nm)	Average D (nm)
Mix + PMMA + Ag NPs	(111)	38.04	19.007	0.2362	35.6	45.6
	(200)	44.27	22.139	0.3936	21. 7	
	(220)	64.75	23.165	0.1181	79.5	
Mix + PMMA + Cu NPs	(111)	43.200	21.6	0.0787	108.5	72.8
	(200)	50.361	25.180	0.2362	37.1	
Mix + PMMA + SiO_2 NPs	(102)	31.592	15.796	0.1181	52	52

(r.m.s.) value of surface roughness and the average diameters increase with thickness when pure and doped PMMA polymer and nanoparticles are combined to create thin films [22].

At room temperature, the absorption spectra of the organic laser dye combinations were recorded using an Aquarius-7000 UV-Visible spectrometer. Fig. 7. depicts the linear absorption spectra of PMMA and nanoparticle-doped dye thin films at a concentration of 10^{-3} M. According to these findings, pure mixed dye, including PMMA polymer and silver nanoparticles, copper nanoparticles, and silicon nanoparticles, caused the absorption peaks to move to longer wavelengths. Electrons in these states go from the Lowest-Occupied Molecular Orbital (LUMO) to the Highest-Occupied Molecular Orbital (HOMO). The linear absorption coefficient and linear refractive coefficient for each sample are calculated from Equations 1 and 2, as illustrated in Table 2. The present results show that the mixture's highest absorption wavelength is 540 nm [23].

Utilizing the Z-scan method to open and closed aperture produces the nonlinear refractive index and nonlinear absorption coefficient, respectively. A Z-scan measurement was performed on the films prepared by depositing a mixture of organic dyes with a polymer, doped with silver, copper, and silicon dioxide nanoparticles. The nonlinear impact area varies from -2 to 2 millimeters. The

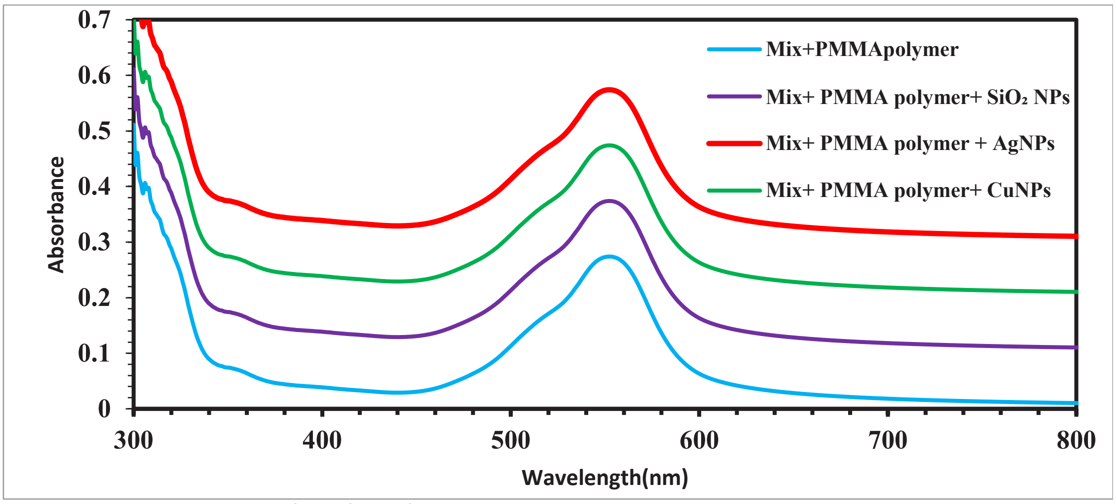

Fig. 7. The absorbance spectra of thin films of organic laser dye doped with PMMA polymer and silver nanoparticles, copper nanoparticles and silicon nanoparticles.

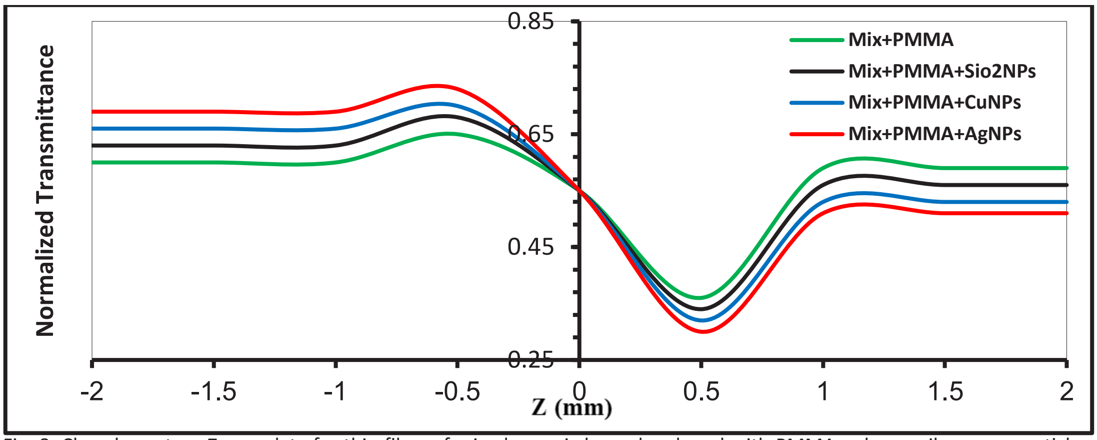

Fig. 8. Closed-aperture Z-scan data for thin films of mixed organic laser dye doped with PMMA polymer silver nanoparticles, copper nanoparticles and silicon nanoparticles.

specimen may feature self-defocus lensing due to a negative sign of refraction nonlinearity (n_2<0), as suggested by the transmittance curve of the Z-scan data for closed apertures, which presents a peak followed by a valley [24]. Figs. 8 and 9 depict the saturable absorption phenomena for combination organic laser dye thin films using open-aperture Z-scan data.

The Z-scan method was employed to study the optical limiting behavior of Fl, E-Y, and RB

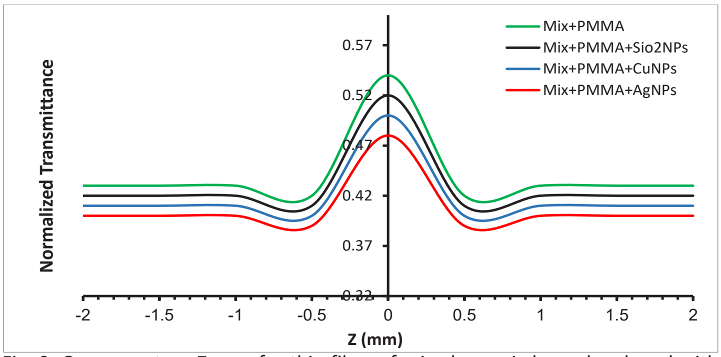

Fig. 9. Open-aperture Z-scan for thin films of mixed organic laser dye doped with PMMA polymer silver nanoparticles, copper nanoparticles, and silicon nanoparticles.

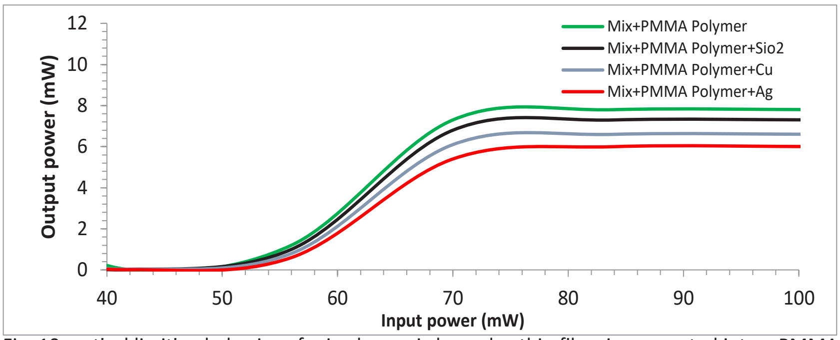

Fig. 10. optical limiting behavior of mixed organic laser dye thin films incorporated into a PMMA polymer matrix and doped with AgNPs, CuNPs and SiO_2NPs.

Table 2. AFM measurements of a combination of PMMA polymer thin films doped with Ag, Cu and SiO_2 nanoparticles.

Sample	Roughness r.m.s (nm)	Average Roughness (nm)	Average Diameter (nm)	Thickness (nm)
Mix + PMMA	3.874	3.182	16.48	124
Mix + PMMA + Sio_2 NPs	5.263	3.906	38.99	133
Mix + PMMA + Cu NPs	5.465	3.928	40.48	141
Mix + PMMA + Ag NPs	8.182	6.906	49.87	161

Table 3. The optical limiting results, linear and nonlinear optical parameters for thin films of PMMA polymer doped with Ag, Cu and SiO_2 NPs at λ=457 nm.

Sample	T	$\alpha_\circ$ (cm^{-1})	$n_\circ$	n_2 (cm^2/mW)10^{-7}	β (cm/mW)	Limiting Threshold (mW)	Limiting Amplitude (mW)
Mix + PMMA	0.9804	1605.546748	1.040087914	10.01×10^{-6}	0.455	7.8	72.5
Mix + PMMA + Sio_2 NPs	0.8916	9326.681911	1.25050274	15.22×10^{-6}	2.377	7.3	72.3
Mix + PMMA + Cu NPs	0.7957	18583.15041	1.546584301	20.70×10^{6}	4.290	6.6	71.4
Mix + PMMA + Ag NPs	0.734	25145.76423	1.790611879	27.75×10^{-6}	4.879	6	70.5

mixture organic laser dyes fabricated as PMMA-based thin films and doped with AgNPs, CuNPs, and SiO_2NPs at a concentration of 10^{-3} M. The resulting optical limiting properties, as well as the corresponding linear and nonlinear optical parameters, are presented in Table 3. Similar to polymer- and nanoparticle-treated dyes, the inverse relationship between concentration and optical power limiting threshold indicates that the optical limiting performance is enhanced at higher concentrations, as illustrated in Fig. 10. This behavior is primarily governed by the optical limiting threshold intensity of each sample [25]. In comparison with solution-based materials Fl, E-Y and RB mixture dye thin films exhibit a lower optical limiting threshold.

CONCLUSION

In this study, nonlinear optical properties of a mixture of (Fl, EO-Y and RB) Organic Laser Dyes as thin films, the absorbance rose along with the concentrations of nanoparticles. Each sample's closed aperture produces self-defocusing occurrences. Every thin-film sample has an open aperture that provides saturable absorption. It is found that all thin films exhibit highly nonlinear characteristics, with a linear increase in the nonlinear refractive index and nonlinear phase shift. Because the combination of laser dyes doped with polymer PMMA and silver nanoparticles exhibits superior optical limiting qualities compared to thin films doped with copper and silicon nanoparticles. They may be utilized as more effective optical limiting in electro-optical systems. The findings also suggest that all samples show promise for use as active laser media and as media for other optoelectronic applications.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this manuscript.

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