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Original Research Article

Nonlinear absorption in dye doped polymer matrices measured by open aperture Z-scan studies

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ABSTRACT
We present the findings of open aperture Z-scan investigations done on Coumarin 540 doped polymer thin films in this work. We looked at the nonlinear absorption in thin films made of dye-doped polymethyl methacrylate (PMMA), polystyrene, and polyvinyl chloride (PVC). Due to reversible saturable absorption, all of the polymer thin films displayed good nonlinear characteristics. The investigations employ the open aperture Z-scan method. The open aperture Z-scan measurements reveal a sample's nonlinear absorption. A transmission minimum at the focus point is observed in the experiments if nonlinear absorption, such as two-photon absorption, is present. When a sample is a saturable absorber, transmission rises with the intensity of the incident light and reaches a maximum at the focal point.

1. Introduction

Nonlinear optical characteristics of materials have grown in significance since the development of the laser. Due to third order nonlinearity, a medium's refractive and absorptive characteristics change when a strong laser beam passes across it. Due to the rising interest in the applications of nonlinear optical characteristics for optoelectronics, different nonlinear optical devices, optical switching, optical limiting, etc., several research have been conducted on the study of these features [1, 2]. Additionally to well-known nonlinear materials as porphyrins, pthalocyanine, and fullerenes, organic dyes are discovered to be potential nonlinear materials. Research is being done to examine the nonlinear optical characteristics of dyes that exhibit large excited state and two-photon absorption (TPA) cross sections [3, 4].

Saturable absorption (SA) and reverse saturable absorption are the two forms of nonlinear absorption seen in dye molecules (RSA) If the excited states exhibit rising absorption saturation due to their lengthy lifetime, the transmission will exhibit SA characteristics. RSA features can be seen in transmission if the excited state exhibits high absorption relative to the ground state. While dyes with RSA properties are employed for optical limiting devices, those with SA qualities are frequently used in mode-locking.

When an intense beam of light travels through a medium, a common approach for evaluating optical nonlinearities of materials is the Z-scan technique. To assess the sign and magnitude of both the real and imaginary parts of third order nonlinear susceptibility, this straightforward and sensitive approach uses a single beam [5]. Along the single beam arrangement, the sample's transmittance is assessed as it travels in the path of a focussed Gaussian laser beam. The method is referred to as closed aperture Z-scan when the transmitted light is measured through an aperture that is positioned in the far field relative to the focus region. Here, nonlinear refraction and absorption of nonlinear energy can affect the transmitted light. On the other hand, open aperture Z-scan is the measurement technique used when all of the light is collected without the use of an aperture. The output is then only responsive to nonlinear absorption.

The open aperture Z-scan technique is employed in the current study to conduct the investigations. The open aperture Z-scan measurements reveal a sample's nonlinear absorption. A transmission minimum at the focus point is observed in the experiments if nonlinear absorption, such as two-photon absorption, is present. When a sample is a saturable absorber, transmission rises with the intensity of the incident light and reaches a maximum at the focal point. We present the findings of open aperture Z-scan investigations done on Coumarin 540 doped polymer thin films in this work. We have investigated the nonlinear absorption in dye doped polymethyl methacrylate (PMMA), polystyrene and polyvinyl chloride (PVC) thin films. All the polymer thin films exhibited good non linear properties due to reversible saturable absorption.

2. Experimental procedure

Coumarin 540 dye is mixed with various polymers, such as PMMA, polystyrene, and PVC, to create thin polymer films that are solid state dye doped. The solvents used for PMMA, polystyrene, and PVC are methyl ethyl ketone, toluene, and cyclohexanone, respectively. For each polymer, a particular weight percentage is used to provide the ideal viscosity for the production of high-quality films. The dye concentration is fixed as 5×10^{-4} M. By using a free cast evaporation technique, the films are created on microslides. The films range from 30

to 300 micrometres in thickness. This study's laser source is a Quanta Ray MOPO (MOPO 700) that is pumped by a Q-switched Nd:YAG laser at 355 nm and emits pulses with a 5 ns duration and a 10 Hz repetition rate. Before accessing the sample, a little portion of this laser's output is made to pass via an achromatic convex lens. At 590 nm, the lens generates a beam waist radius of 39 micrometres. The samples with thickness less than 300 micrometres satisfy the narrow lens approximation since the associated Rayleigh range Z0 is 8.2 mm. This will prevent the beam diameter from changing at the sample's entry and exit sides [5]. A motorised translation stage moves the sample through the beam waist while an energy ratio-meter from Laser Probe Inc. equipped with RjP 735 probes measures the ratio of transmitted and incident energies. Using LabView, the entire experimental setup is automated. Near the resonance wavelength of 590 nm and an off resonance wavelength of 650 nm, where the absorption is minimal, the sample's nonlinear absorption is investigated.

3. Results and discussion

Using a UV-VIS spectrophotometer, the linear absorption spectra of dye-doped films are captured (JascoV570). Figure 1 shows the dye-doped PMMA absorption spectrum. The absorption spectra of the other polymer matrices shared a similar characteristic. In dye-doped PMMA, polystyrene, and PVC polymer matrices, open aperture Z-scan investigations are carried out for varied input intensities. Excitation wavelengths of 590 nm at near resonance and 650 nm at off resonance were used to investigate the nonlinear absorption in the dye doped materials. Figure 2 displays typical C-540 dye doped PMMA thin film open aperture Z-scan curves with normalised transmittance.

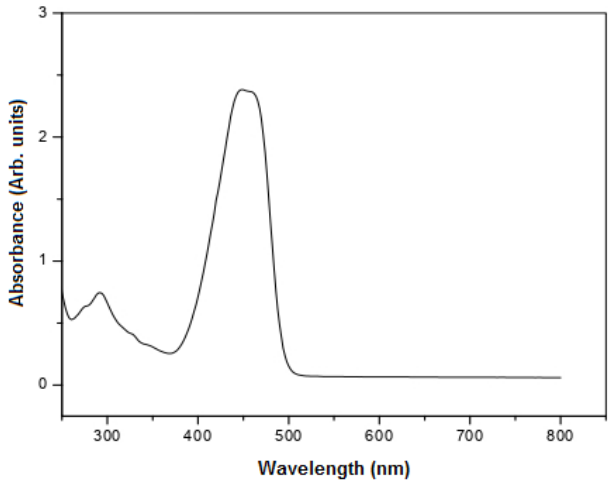

Figure 1. Absorption spectrum for C-540 doped PMMA thin film.

The transmittance is minimum at the focus and increases steadily on both sides of the focus. The Z-scan measurements for the different polymer matrices exhibited RSA behavior for different pump intensities. From the Z-scan plots, the nonlinear absorption coefficient β is obtained by fitting the experimental open – aperture z-scan plots to the equation:

$$T_{(Z)} = \frac{C}{q_0\sqrt{\pi}} \int_{-\alpha}^{\alpha} \ln(1+q_0 e^{-t^2})dt \,,$$

where $q_0 = \frac{\beta I_0 L_{eff}}{1+x^2}$.

Here C is the normalizing constant, I_0 is the irradiance at focus, $x = z/z_0$.

$L_{eff} = \frac{1-e^{\alpha_0 l}}{\alpha_0}$ is the effective length and l is the sample length [5].

β is usually called β_{eff} which takes into account all the possible nonlinear absorption mechanisms present in the sample. β_{eff} is a measure of the strength of nonlinear absorption and optical limiting. The measured values of β_{eff} and irradiance I_0 for different dye doped polymer matrices are given in Table 1. These measurements are done at an excitation wavelength of 590 nm. From the table, it is clear that is higher for dye doped polystyrene films comparing to the other two polymer matrices. The Z-scan plots for various irradiances are shown in Figures 2 and 3 for PMMA and polystyrene matrices, respectively.

Table 1: Values of irradiance and β_{eff} for different dye doped polymer matrices.

Sample	Thickness of film (μm)	Irradiance (MW cm^{-2})	β_{eff} (cm M W^{-1})
Dye doped PMMA	190	746	0.047
Dye doped polystyrene	30	400	6.34
Dye doped PVC	14	573	2.167

Z-scan measurements are repeated at an off resonance wavelength of 650 nm. The nature of the Z-scan plots is similar to that at resonance wavelength. The values are observed to be comparatively less.

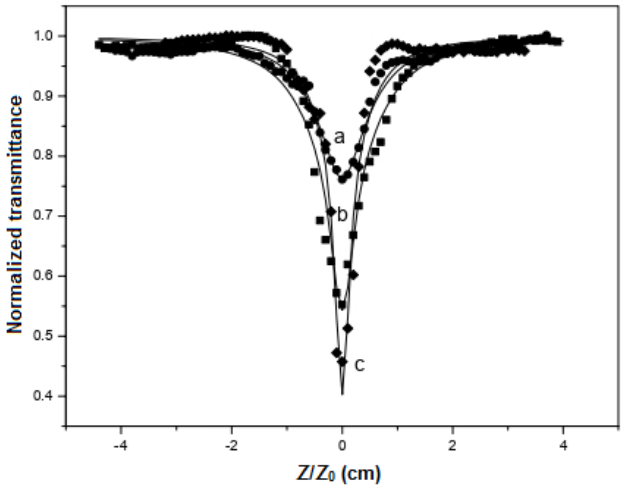

Figure 2. Open aperture Z-scan curves for C-540 doped PMMA films for different irradiances. (a) 805 MW cm^{-2}, (b) 985 MW cm^{-2}, (c) 1850 MW cm^{-2}.

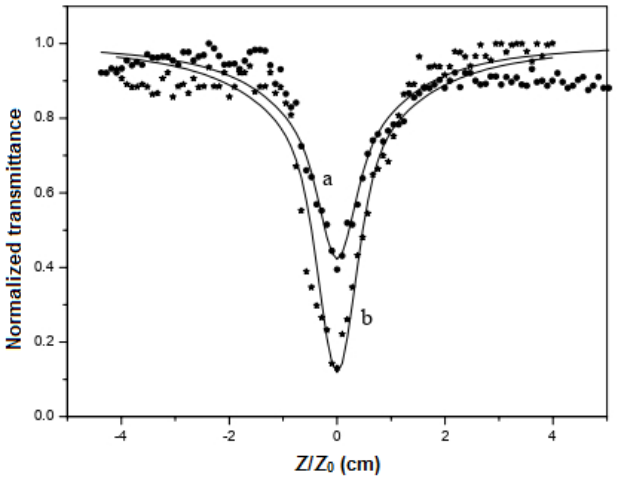

Figure 3. Open aperture Z-scan curves for C 540 doped polystyrene films for different irradiances. (a) 322 MW cm^{-2}, (b) 530 MW cm^{-2}.

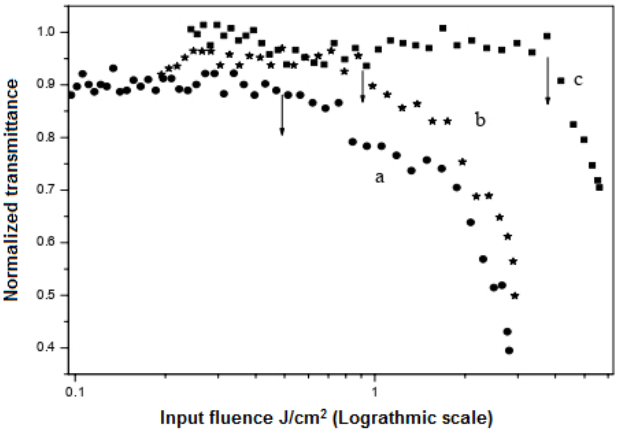

Figure 4. Nonlinear transmission in dye doped polymer films. (a) polystyrene, (b) PMMA, (c) PVC.

We can plot nonlinear transmission as a function of input fluence to compare the impact of intensity on nonlinear absorption of various polymer matrices. These charts are produced using the Z-scan data. Fluence levels at different sample points can be determined using the Gaussian beam waist standard equation from the value of fluence at the focus. The nonlinear transmission for the various dye-doped polymer samples is shown in Figure 4. The arrow shows around what fluence level the transmission departs from linear behaviour. The figure demonstrates that dye doped polystyrene films have a higher optical limiting property.

4. Conclusions

All of the charts generated for the open aperture Z-scan data were deemed to be typical of ESA [6] based on the study. The effect of TPA in nonlinear absorption may not be important because there is no discernible difference between Z-scan plots near resonance and off resonance wavelengths. TPA's nonlinear absorption may cause fluorescence emission because it is a very fluorescent substance. During the Z-scan measurements, no such fluorescence emission is seen. The current open aperture Z-scan measurements highlight the intriguing characteristic of Coumarin 540 doped polymer films' nonlinear absorption and imply that excited state absorption may be involved in the nonlinear behaviour.

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