MEASUREMENT OF NONLINEAR RESPONSES AND OPTICAL LIMITING BEHAVIOR OF TiO_2/PS NANO-COMPOSITE BY SINGLE BEAM TECHNIQUE WITH DIFFERENT INCIDENT INTENSITIES

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Titanium dioxide (titania) is a cheap, nontoxic and highly efficient photocatalyst being extensively applied for the degradation of organic pollutants, air purification, water splitting, reduction of nitrogen to ammonia, and optical devices due to its optical behaviors. The third-order nonlinear optical properties of TiO_2/PS nano-composite were studied by means of single beam transmission technique, using a continuous-wave (CW) He–Ne laser beam with a wavelength of 632.8 nm with three different incident intensities. The magnitude and sign of the third-order nonlinear refractive index (n_2) and nonlinear absorption (β) of TiO_2/PS nano-composite were determined by use of both the closed-aperture and opened-aperture z-scan techniques. Z-scan technique, developed by Sheik-Bahae et al., has been used widely in material characterization. A single-beam (also called z-scan method) for measuring the sign and magnitude of nonlinear refraction that has simplicity and very high sensitivity has been reported recently. Optical limiting property of TiO_2/PS nano-composite is studied.

The positive sign obtained for nonlinear refractive index indicated that there is a self-focusing effect in the sample. The nonlinear refractive index was in order of 10^{-8} (cm^2/W) and the nonlinear absorption coefficient was obtained in order of 10^{-2} (cm/W) with negative sign. The values of nonlinear refractive index and nonlinear absorption coefficient are enhanced by decreasing the intensity.

Keywords: TiO_2/PS nano-composite; z-scan technique; Nonlinear refraction; nonlinear absorption.

1. Introduction

An important feature of nano-particles is to have very large ratio of surface atoms to volume atoms. The existence of this vast surface can have a profound effect on the physical and chemical properties of nano-particles, and these properties are also intensely affected by surrounding medium.¹ In the quantum-confined regime, nano-particles exhibit novel physical and chemical properties, especially nonlinear optical properties.²

Recently, a large number of photonic applications such as optical switches and optical adulators, based on the nonlinear optical properties of different materials have been developed. The nonlinear optical properties of TiO₂/PS nano-composite have been investigated by single-beam transmission technique. Z-scan is a versatile method that has been widely employed for determining the non-linear absorption and refraction of a host of materials.³

Therefore, these nano-materials are very useful in applications in optical limiting and NLO devices.⁴ In this letter, we report the main optical properties such as the sign and magnitude of both the nonlinear absorption and refractive index and linear absorption of TiO₂/PS nano-composite by using the z-scan technique.

2. Experimental section

To further study the influence of laser intensity on the absorption property, we measured the transmittance of the sample at focal position with different laser intensities. In order to describe the nonlinear optical properties at the colloid at low concentrations it is used an extension of the Maxwell-Gamet model.⁵

The obtained TiO₂/PS nano-composite nonlinearities were aged in darkness at room temperature and have high stability. The beam waists in focal plane were measured to be 34 μm for the wavelengths of 632.8 nm.

2.1. Optical limiting (OL)

Linear optical absorption measurements of the samples were performed in the air at room temperature by optical limiting (OL) experiment. We calculate the linear absorption, α , by measuring output power with a sample as a function of the power without the sample in low powers. The linear absorption is obtained from Eq. (1).

$$\alpha = -\frac{1}{L} \ln\left(\frac{P}{P_0}\right) \quad (1)$$

2.2. Z-scan technique

The experimental z-scan setup is shown in figure1. Here in z-scan experiment, at each position (Z) the sample sees different laser intensity, and the position dependent (ie, intensity-dependent) transmission is measured using a power meter's detector placed after the sample. The transmissions with and without the aperture were measured in the far field as the sample was moved through the focal point, this enables the separation of the nonlinear refractive index from nonlinear absorption.⁶

Fig.1. Experimental close aperture z-scan setup.

2.3. Close aperture z-scan

The nonlinear refractive index, n_2 in the expression was calculated as follows,

$$n(I) = n_0 + n_2 I \quad (2)$$

Where n_0 is the linear refractive index and I is the intensity of the incident laser light.⁷ The z-scan measurement with an aperture (close aperture) was also performed for the investigation of nonlinear refraction of TiO₂/PS nano-composite.⁸ Typical peak – valley (valley – peak) transmittance curve is obtained when the nonlinear refractive index of the medium is negative (positive).⁹ By monitoring the transmittance change through a small circular aperture placed at the far-field position, one is able to determine the nonlinear refractive index.¹⁰ The samples were moved in step of 1 mm during the scan.

The relation between the normalized transmittance $T(z)$ and z position was obtained by moving the samples along the axis of the incident beam (z -direction) with respect to the focal point. The variation of this quantity as a function of $\Delta\phi_0$ is given by

$$\Delta T_{p-v} = 0.406(1-S)^{0.25} \Delta\phi_0 \quad (3)$$

Where S is the linear transmittance of the far-field aperture. $|\Delta\phi_0|$ relates to n_2 through the following expression,

$$\Delta\phi_0 = k L_{\text{eff}} n_2 I_0 \quad (4)$$

Where I_0 is the intensity of the laser beam at focus $z=0$, $L_{\text{eff}} = \frac{1 - \exp^{-\alpha L}}{\alpha}$ is the effective thickness of the sample, α is the linear absorption coefficient and L is the thickness of the sample. The nonlinear refractive index n_2 (cm²/w) can be obtained from Eqs. (3) and (4).¹¹

2.4. Open aperture z-scan

We deduce the nonlinear absorption coefficient, β in the following expression for the absorption from the nonlinear transmission measurements,⁷

$$\alpha(I) = \alpha_0 + \beta I \quad (5)$$

In a typical open aperture z-scan, the open aperture transmittance normalized to the linear transmission of the sample is plotted against the sample position measured relative to the beam focus. Nonlinear absorption is usually indicated by a smooth valley-shaped curve which is symmetric about the focal point ($z = 0$).¹² Considering the two-photon absorption, the normalized transmittance $T(z)$ for the open aperture z-scan can be written as follows:

$$T(z) = \sum_{m=0}^{\infty} \frac{[-q_0(z,0)]^m}{(m+1)^{\frac{3}{2}}} \quad (6)$$

Where $q = \beta I_L \frac{(1 - e^{-\alpha L})}{\alpha}$. Also β is the two-photon absorption coefficient, $I_0(t)$ is the on-axis instantaneous intensity of the laser beam at the focus (z_0), $z_0 = \frac{\pi \omega_0^2}{\lambda}$ is the diffraction length of the beam, and z is the sample position.¹³

The materials exhibit both nonlinear refraction and absorption properties; we can obtain pure nonlinear refractive index n_2 by dividing the closed aperture data by the open aperture data.¹¹ The nonlinear optical responses of the TiO_2/PS nano-composite were investigated using the above-described z-scan system.

3. Results and discussion

In our experiments, all z-scans performed at different incident laser intensities display a decrease in transmittance at the focus, typical of an induced negative NLO absorption of incident light.

3.1. Result of OL experiment

In the OL experiments, these samples were placed at focus of a lens with 8 cm focal length. Both the incident and transmitted laser power was monitored using power meter's detector. Experimental data is shown in figure2. The linear absorption was found to be about $\alpha = 2.72 \text{ cm}^{-1}$.

Fig. 2. Optical limiting behavior of TiO₂/PS nano-composite.

3.2. Result of close aperture z-scan

The close-aperture (CA) curve was obtained after division of CA data with the OA data to eliminate the contribution of nonlinear absorption and is shown in figure 3. The difference between normalized peak and valley transmittance ΔT_{p-v} (denoting $T_p - T_v$) can be directly measured by z-scan technique. The nonlinear refractive index n_2 (cm²/w) can be obtained from Eqs. (3) and (4). The peak-to-valley configuration of the CA/OA z-scan curve for TiO₂/PS nano-composite reveals a self-focusing effect, and the refractive index has a positive sign.

Fig. 3. Z-scan data of TiO₂/PS nano-composite with a closed-aperture at different incident intensities.

3.3. Result of open aperture z-scan

We use the open aperture z-scan to determine the β value of nano-particles.¹⁴ The open circles in figure 4 are measured without the aperture at three input intensities. The solid curve in figure 4 is fitted by using Eq. (6) with the parameters in our experiment and it can be seen that the theoretical curve fits well with the experimental data.

Fig. 4. Open aperture z-scan measurements on TiO₂/PS nano-composite at three input intensities (Normalized Transmittance versus distance, z.).

The measured nonlinear refraction values, n_2 , and nonlinear absorption values, β , at three incident intensities are summarized in Table 1.

Table 1. Calculated values of linear absorption coefficient, nonlinear absorption coefficient and nonlinear refractive index at three incident intensities.

$I_0(\text{kW}/\text{cm}^2)$	$\alpha (\text{cm}^{-1})$	$\Delta\Phi_0$	$n_2(\text{cm}^2/\text{W})$	$\beta(\text{cm}/\text{W})$	$P_0(\text{mW})$
2.754	2.72	0.38	1.6×10^{-8}	1.05×10^{-2}	50
1.377	2.72	0.56	4.7×10^{-8}	4.03×10^{-2}	25
0.826	2.72	0.64	9×10^{-8}	6.9×10^{-2}	15

4. Conclusion

In this paper, refractive and absorptive nonlinearities of TiO₂/PS nano-composite were observed in z-scan experiment under CW He-Ne laser. The close aperture z-scan measurements at 632.8 nm revealed that the nonlinear refractive index in TiO₂/PS nano-composite is in the range of $10^{-8} \text{ cm}^2/\text{W}$. The positive sign of n_2 showed the occurrence of self-focusing phenomena is take placed. A nonlinear absorption (NLA) was detected this was mainly associated with two-photon absorption (TPA) effect. Their nonlinear optical absorptions reached as $10^{-2} \text{ cm}/\text{W}$ measured using open aperture z-scan measurement.

It was also found that the nonlinear refractive index and nonlinear absorption values of TiO₂/PS nano-composite decreases with increasing incident intensity. The large optical nonlinearity with ultrafast response of TiO₂/PS nano-composite indicated their potential applications for future nonlinear optical devices.

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