



ENHANCING THE NONLINEAR OPTICAL PROPERTIES OF ORGANIC DYE BY USING NANOPARTICLE COMPOUNDS

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ABSTRACT

Nonlinear optical properties of Rhodamine 6G dye doped epoxy resin with Al_2O_3 nanoparticle at different concentration were studied by using EZ-scan technique to evaluate the nonlinear parameters such as nonlinear absorption coefficient (β), nonlinear index of refraction (n_2) and the third order optical nonlinearity. The samples with the nanoparticle exhibit negative sign of the nonlinear refractive index n_2 indicates that the samples exhibit self-defocusing optical nonlinearity. The samples with nanoparticle showed reverse saturation absorption (RSA) in open EZ-scan. The nonlinear refractive index n_2 , the nonlinear absorption coefficient β and the third-order optical susceptibility $\chi^{(3)}$ values are of the order of $10^{-9} \text{ cm}^2/\text{W}$, $(1-4) \text{ cm/W}$ and $10^{-6} \text{ V}^2/\text{m}^2$ respectively.

Keywords: Rhodamine 6G, EZ-scan, epoxy resin, nanoparticle.

1. INTRODUCTION

Rhodamine 6G (R6G) is one most important of Rhodamine dyes in xanthine family dye, which emits laser light of a broad range of wavelength in the visible region of the spectrum. R6G dye usually pumping at a wavelength (532) nm. Also, it has highly efficient, where that up to 20% of the input energy is converted to laser light output, and the fluorescence lifetime about (4) nsec [1]. The dye in liquid state has some problems such as, a fast photo-degradation, unstable and difficult to use in the devices, therefore, the use of the polymer host presents some features such as toxicity removed, flammability, and easy to operate and maintain [2,3,4,5].

Nonlinear optics (NO) is the study of optical phenomena that get as a consequence of the modification of the medium by the presence of light. Typically, the laser light can modify the properties of a material [6]. Organic materials consider one of the important nonlinear optical materials because their properties such as, exhibit large nonlinear properties, fast nonlinearities response, easy to process and integrated into optical devices [7].

There has been considerable scientific research on the development of nonlinear optical materials with large third-order nonlinear susceptibility for photonic devices. In this direction, conjugated polymers have been investigated extensively due to their inherently large ultrafast resonant nonlinearity for the improvement by way of molecular engineering [8]. Also, improvement nonlinear optical properties of polymer samples by using nanoparticle.

In this work, nonlinear optical properties of Rhodamine 6G (R6G) doped epoxy resin were studied, with take effect the concentration of dye nanoparticle and nanoparticle (Al_2O_3) at different weight (0.05, 0.1 and 0.15 %), and thickness (0.5) mm, by using EZ-scan method.

2. EXPERIMENTAL

2.1 Material

The dye which has been used in the study is Rhodamine 6G (R6G) supplied by the HiMedia Laboratories Company. The dye is powder solid dark reddish purple, the molecular formula ($\text{C}_{28}\text{H}_{31}\text{N}_2\text{O}_3\text{Cl}$), molar mass (479.02 g/mol), and the structure of R6G dye shown in Figure-1. The dye doped in epoxy resin (Euxit 50 KI) supplied by Egyptian swiss chemical industries company, which consists of two materials, the first called the base (resin), denoted by the letter (A), and the second material called sclerosing (hardener) and denoted by the letter (B) with ratio mixer (A:B) (3:1). Addition to Al_2O_3 nanoparticle with particle size (20-30) nm, and which supplied from Intelligent Materials Pvt. Ltd Company.

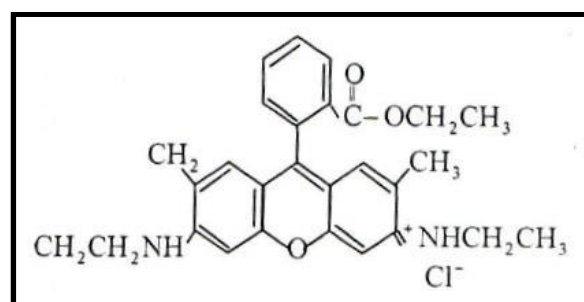


Figure-1. The structure of Rhodamine 6G dye.

2.2 Eclipsing Z-scan

The nonlinear optical properties of dyes were evaluated by employing the EZ-scan technique developed by T. Xia *et al.* EZ-scan experiments were performed with Nd:YAG laser at (532) nm wavelength. The laser beam has a beam waist of (85) μm and the Rayleigh length Z_R of (42) mm. A 30 cm focal length lens was used with input power (1) mW, and the sample thickness is (0.5) mm. Hence, the sample is thin because the Rayleigh length Z_R larger than of the sample thickness.

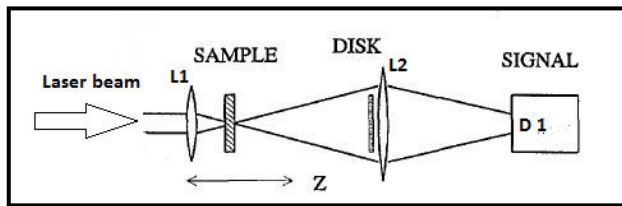


Figure-2. Experimental arrangement for the EZ-scan.

The nonlinear absorption coefficient was calculated by fitting the normalized transmittance $T(z)$ [9]:

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

Where: $q_0(z) = I_0 L_{eff} \beta / [1 + (z/z_0)^2]$, and $L_{eff} = \frac{1 - e^{-\alpha_0 L}}{\alpha_0}$, the effective length of the sample, L is the thickness of the sample, α is a linear absorption coefficient, and I_0 is the intensity of the laser beam at the focus ($z = 0$). We have chosen β as a free parameter. The β value can then be extracted from the best fitting. The nonlinear refractive index n_2 was calculated according equation [10]:

$$n_2 = \Delta\Phi_0 / I_0 L_{eff} k \quad (2)$$

$$\text{Where: } \Delta T_{pv} = 0.68(1 - S)^{-0.44} |\Delta\Phi_0|.$$

3. RESULTS

3.1. Absorption spectrum

The absorption spectra of the R6G dye are shown in figure (3). The absorption of the samples varying with wavelength at range (40-700) nm, and the peaks is broad in the visible region. The absorption at around 500 to 600 nm can be due to the $\pi-\pi^*$ transition.

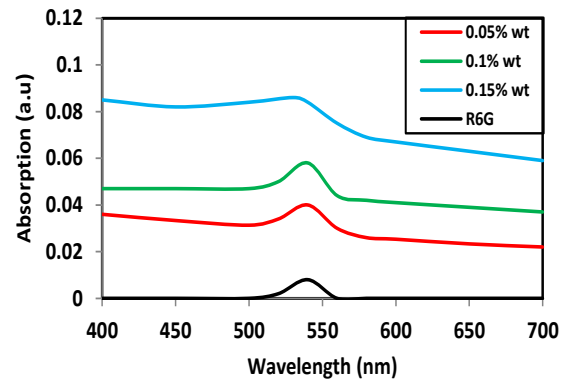


Figure-3. Absorption spectrum of the samples doped Al_3O_2 nanoparticle.

The peaks of samples at a wavelength (540) nm, and shifted to shorter wavelength (535) nm with increases the nanoparticle concentration.

3.2 Nonlinear optical properties

Open EZ-scan aperture experiments have been used in order to calculate the nonlinear absorption coefficient β of the R6G dyes. The transmittance of laser beam after pass through the sample are shown in figure (4), in which the transmittance varies with the position of the sample (z). When the samples are at far field, the transmittance of the laser beam is low, and when the sample close to the focus ($z=0$), the transmittance decreases. It was noted that the decrease in transmittance at the focus ($z=0$) to form a valley, which indicates the reverse saturable absorption (RSA), i.e. positive value for all samples. The RSA behavior can be due to some process such as, excited state absorption (ESA), two-photon absorption (TPA), nonlinear scattering or with these processes [11].

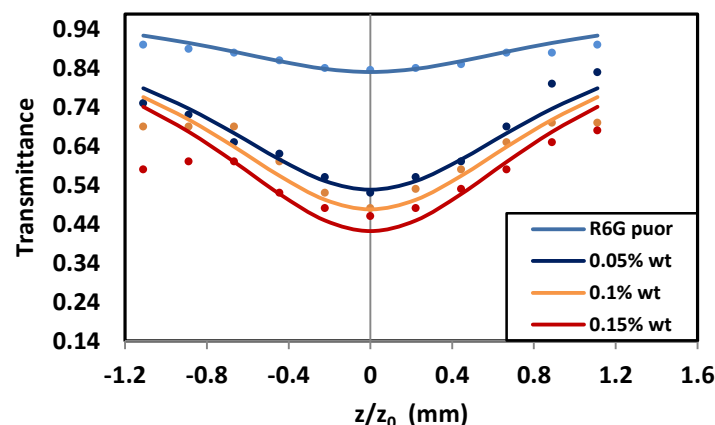


Figure-4. Open EZ-scan of solid samples doped nanoparticle.

The values of the nonlinear absorption coefficient (β) an increase as increase the concentration of the

nanoparticle, as shown in the Table-1. Thus, increase the concentration of the nanoparticle enhancing the nonlinear



absorption coefficient for the R6G dye doped epoxy risen to large value about (4) W/cm, although the value of the power of the laser is very small about (1) mW. This good, to use the properties of these materials in various optical applications.

The closed aperture EZ-scan was used by placing the disc in front of the photodetector, this allowed to determine the magnitude and sign of the nonlinear refractive index (n_2) of the dyes. The closed aperture curve with close EZ-scan of the R6G dye at different concentration of nanoparticle is shown in figure (5). The EZ-scan curve exhibits peak-valley and change to valley-

peak with addition the nanoparticle, this signature indicates the self-defocusing and self-focusing, where self-defocusing is negative nonlinear refractive index n_2 and self-focusing is positive nonlinear refractive index n_2 [12].

The solid sample doped nanoparticle contribute in enhancing nonlinear refractive index of samples, where the values of the nonlinear refractive index of samples with nanoparticle are the larger of the samples without nanoparticle, and the values of the nonlinear refractive index (n_2) increasing as the nanoparticle weight increases as shown in Table-1.

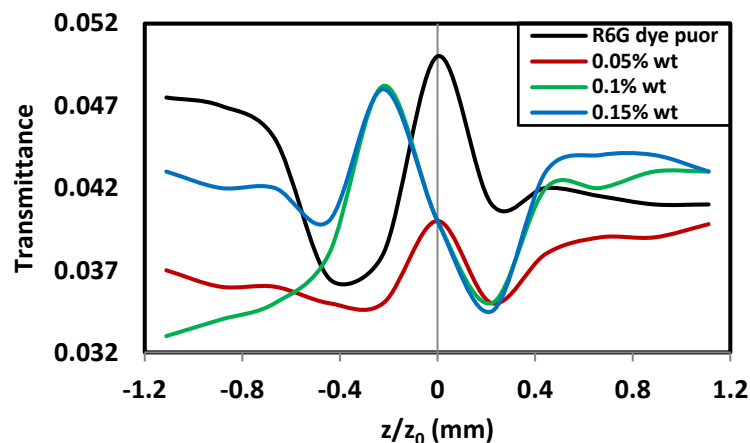


Figure-5. Close EZ-scan of solid samples doped nanoparticle.

The third-order nonlinear susceptibility $\chi^{(3)}$ is dependent on the nonlinear refractive index (n_2) and nonlinear absorption coefficient (β), which determined according to equations 3,4 and 5 [13]:

$$\chi^{(3)} = \chi_R^{(3)} + \chi_I^{(3)} i \quad (3)$$

The imaginary part is related to the nonlinear absorption coefficient β through:

$$\chi_I^{(3)} = \frac{n_0^2 \epsilon_0 c \lambda}{2\pi} \beta \left(\frac{\text{m}^2}{\text{V}^2} \right) \quad (4)$$

The real part is:

$$\chi_R^{(3)} = 2n_0^2 \epsilon_0 c n_2 \left(\frac{\text{m}^2}{\text{V}^2} \right) \quad (5)$$

The absolute value of third order nonlinear susceptibility is:

$$|\chi^{(3)}| = \sqrt{(\chi_I^{(3)})^2 + (\chi_R^{(3)})^2} \left(\frac{\text{m}^2}{\text{V}^2} \right) \quad (6)$$

Where:

- n_0 : Linear refractive index,
- ϵ_0 : Vacuum permittivity (8.85×10^{-14} F.cm⁻¹).
- c : Light speed in vacuum (3×10^{10} cm/sec).
- n_2 : Nonlinear refractive index (cm²/W).
- β : Nonlinear absorption coefficient (cm/W).
- λ : The wavelength (cm).

Table-1. Nonlinear properties of samples doped nanoparticle.

Wight ratio %	n_2 (cm ² /W) 10^{-9}	β (cm/W)	$ \chi^{(3)} $ (V ² /m ²) 10^{-6}
0.0	0.14	1.1	0.025
0.05	0.054	3.2	0.39
0.1	0.143	3.55	0.6
0.15	0.15	4.05	0.61



The nonlinear refraction index n_2 , nonlinear absorption coefficient β and the third order nonlinear susceptibility $|\chi^{(3)}|$ increase linearly with the increase of nanoparticle concentration in samples, this may be attributed to increase the particles which absorb the laser, and enhancing the thermal effect, where, the nonlinearity is due to thermal effects under CW laser irradiation [14]. The results of nonlinear optical properties of R6G dye doped epoxy resin with Al_2O_3 nanoparticle are agree with the work of researcher [15].

4. CONCLUSIONS

We have studied the nonlinear optical properties of R6G dye doped epoxy resin at various concentrations of Al_2O_3 nanoparticle by using EZ-scan method with Nd:YAG laser at 523 nm wavelength. The dyes were characterized with negative nonlinear refraction without nanoparticle and shifted to positive nonlinear refraction with nanoparticle. It was observed an increase in nonlinear optical properties of samples with increase in concentration of nanoparticle, and the samples showed large nonlinearity properties.

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