IOSR Journal Of Applied Physics (IOSR-JAP)
e-ISSN: 2278-4861.Volume 12, Issue 2 Ser. I (Mar. – Apr. 2020), PP 20-25
www.Iosrjournals.Org

Investigation Of Nonlinear Refractive Index For 2,5-Dimethaylaniline (DMA) Under Different Polarized Light

[1]Nujhat Nuri Sultana, [2]Nusrath Ruhaney, [1]Ponkog Kumar Das, [1]Md. Enamul Hoque, [1]Manash Kanti Biswas, [1]Sharif Md Sharafuddin,[1]Yasmeen Haque

[1]Nonlinear Optics Research Laboratory, Department of Physics, Shahjalal University of Science and Technology, Sylhet-3114, BANGLADESH.
[2]Department of Computer Science and Engineering, Faculty of Agricultural Engineering and Technology, Sylhet Agriculture University, Sylhet-3114, BANGLADESH.

***Abstract**: Lights of different polarizations have been used to measure the value of n_2 by using the polarization changeable Z-scan technique. The nonlinear refractive index n_2 was measured for different concentrations of 2,5-dimethaylaniline in Chloroform. An Ar-ion laser* was *used with varying incident powers for three different wavelengths 457 nm, 488 nm and 514 nm. The results give a negative n_2 with a magnitude of the order of $10^{-6}cm^2/W$. For all the wavelengths, the value of n_2 was found to decrease with different states of lights i.e. linear, elliptical and circular polarized lights respectively, but the order of magnitude of n_2 was the same.*

Keywords: *Polarization of light, Z scan, Nonlinear refractive index.*

Date of Submission: 02-03-2020 Date of Acceptance: 17-03-2020

I. Introduction

Non linear Optics involves the study of the interaction and propagation of high intensity light through matter. The presence of the light induces a change in the refractive index which can be measured experimentally by means of a nonlinearity parameter n_2.

Organic materials yield a vast amount of information about the optical nonlinearity related to the type of bonding in the materials. One of the main areas of research in the nonlinear optical field is the determination of the nonlinearity parameter n_2 of new unknown materials. The optical nonlinearity of organic materials[1] is of prime interest since it provides a vast information related to the interaction of light with these materials. Applications are abundant in photonics and materials science. Many organic materials show third order optical nonlinearity due to the presence of $\pi -$ electrons in their structure[2]. There are different experimental methods for the measurement of third order nonlinearity, such as nonlinear imaging techniques[3], nonlinear ellipse rotation[4,5], degenerate four-wave mixing[6], and Z-scan technique[7]. Among these methods Z-scan technique is a very popular and standard method for determining nonlinear parameters of various materials because of its simplicity, sensitivity, accuracy and the ease of separation between nonlinear refraction(NLR) and nonlinear absorption(NLA). The reliable Z-scan technique was first introduced by Sheik-Bahae *et al.*[7,8]. Since the experimental work of M .Sheik-Bahae *et al*, there have been various theoretical and experimental modifications of this method, such as thick sample Z-scan[9,10], eclipsing Z-scan[11], pump-probe Z-scan[12,13] etc. Among these Z-scan methods, linearly polarized lights has always been used as the incident beam. R. De Salvo *et al* presented the effect of polarization on Z-scan measurements, the work on polarization dependence of nonlinear refractive index in crystals[14]. An extension of the standard Z-scan method for arbitrary polarized incidence light has been reported by Yan *et al.* in 2009 for measurements of third-order nonlinear susceptibility of CS_2 liquid [15]. The theoretical analysis was examined experimentally by studying third-order nonlinear susceptibility of CS_2 liquid. In this experiment, the closed aperature (CA) case of z-scan has been used to analyze the linearly, circularly, elliptically polarized light z-scan data for CS_2 medium. In 2012, Yan *et al*. also presented the polarization characteristics of nonlinear refractive index and nonlinear scattering in touluene, o-dichlorobenzens, N, N-dimethylformamide using the polarized light Z-scan technique with femto-second laser pulses [16]. Both the nonlinear refractive index and nonlinear scattering in the three solvents (i.e. toluene, o-dichlorobenzens, N, N-dimethylformamide) were functions of the polarization states of the incident light. Yan *et al*. used femto-second laser pulses while probing the electronic contribution. But in this article, the thermal effect is being investigated using elliptical and circularly polarized light.

Organic material 2,5-dimethylaniline (DMA) has been chosen as the sample for our research work, which is the building block of Poly 2,5-dimethylaniline (PDMA). Experimental work with PDMA , DMA and Hibiscus Rosa-Sinensis, using the Z-scan technique has recently been reported with an n_2 value of the order of

the 10^{-14} esu[17,18,19] in these experiments linearly polarized light has been used as the incident beam. In this paper, we report on an experimental research of the polarization dependence of nonlinear refractive index of 2,5-dimethylaniline dissolved in chloroform. The normalized transmittance formula of closed-aperture Z-scan is obtained for linearly, elliptically and circularly polarized incident beams. The normalized transmittances of DMA for linearly, elliptically and circularly polarized incidence beams were measured.
In this work, Polarization dependence Z-scan technique has been implemented for the determination of the nonlinear refractive index (n_2) of DMA.

II. Experimental Details

2, 5-Dimethylaniline (DMA) has been used as the sample in this experiment. DMA is an organic compound, a substituted derivative of aniline. Because of the low transparency of DMA it was dissolved in chloroform. Two different concentrations were used in this experiment.

Z-scan technique is a very popular and simple method for determining third-order optical nonlinearity via nonlinear optical refractive index and absorption measurements. This technique introduced by Bahae *et al.* is now popularly known as the z-scan technique as it involves the motion of the sample across the focal point of the laser beam along the direction of propagation (Fig. 1). Assuming a Gaussian beam (TEM_{00}), this experiment allows an intensity scan of the irradiated sample, and provides information about the nonlinearity in the sample. The typical z-scan is performed by translating the sample along the z axis from one side of the focus to the other (Fig.1). This results in changing the spot size of the incident beam on the sample to a minimum at the focus and then increasing again on crossing the focus. Correspondingly, the intensity of incident light increases on approaching the focus until a maximum at the focus is reached and then reduces on moving away from the focus. Thus, the overall purpose of the experiment is to determine the variation in transmission as the incident intensity changes by translation along the z-axis. The change in the transmittance of the focusing Gaussian beam in a medium is recorded as a function of position of the medium.
This transmittance can be related with the nonlinear phase-shift which is related to the nonlinear refractive index.

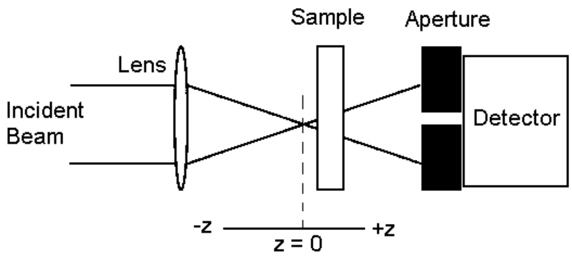

Fig. 1: Schematic diagram of closed aperture Z-scan set up

For a cubic nonlinearity, the refractive index n can be written as [20]

$$n = n_0 + n_2 I \quad (1)$$

Where n_0 is the linear refractive index, I is the irradiance of the laser beam within the sample and n_2 is the nonlinear refractive index.
For a Gaussian beam (TEM_{00}) with beam waist ω_0 moving in the +z direction can be expressed as [21]

$$E(r,z) = E_0 \frac{\omega_0}{\omega(z)} exp\left[-i\left\{kz + tan^{-1}\left({}^{z_0}/_{z}\right)\right\}\right] \quad (2)$$

The parameter is $\omega(z) = \omega_0 \left\{1 + \left({}^{z}/_{z_0}\right)^2\right\}^{1/2}$, ω_0 is the beam waist i.e. , minimum spot size at $z = 0$, $R(z) = z\left\{1 + \left({}^{z}/_{z_0}\right)^2\right\}$ is the radius of curvature of the spherical wave fronts at z , $z_0 = \frac{\pi \omega_0{}^2}{\lambda}$ is the Rayleigh range of the beam, $k = \frac{2\pi}{\lambda}$ is the wave vector, λ is the laser wave length and r (x, y) is the transverse radial distance [7,21,22].

For a thin sample, the far-field condition to give a geometry-independent normalized transmittance is[7,8,23]

$$T(z, \Delta\varphi_0) = 1 + \frac{4x\Delta\varphi_0}{(x^2+1)(x^2+9)} \quad (3)$$

Where $x = z/z_0$.This is the equation for transmittance which is used for theoretical fitting of the closed aperture z-scan technique.
The nonlinear phase shift, $\Delta\varphi_0$is related to the nonlinear refractive index, n_2 by

$$n_2 = \frac{\Delta\varphi_0}{kI_0L_{eff}} \quad (4)$$

Where $L_{eff} = \frac{1-e^{-\alpha L}}{\alpha}$, α is the linear absorption coefficient and L is the sample length.

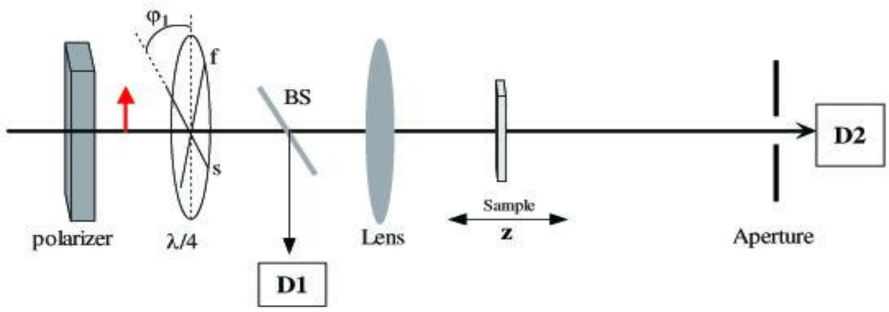

Fig. 2: Experimental arrangements for polarization changeable Z-scan

The modified Z-scan experimental arrangement is shown in Fig.2. Compared with the normal Z scan method, a quarter-wave plate is placed between the polarizer and the sample to create different polarized lights. When a linearly polarized beam passes the quarter-wave plate with angle φ_1 ($-\pi/2 \leq \varphi_1 < \pi/2$) which is the angle between the linear polarization direction and the slow axis of the quarter-wave plate, it can be converted into a polarized beam ($\varphi_1 = -\pi/2$, 0 for linearly polarized beam, $\varphi_1 = \pm\,\pi/4$ for circularly polarized beam, and others for elliptically polarized beam)[15].

A continuous wave (CW) Ar-ion laser source, a polarizer, a quarter-wave plate and a convex lens (focal length 5.02cm) were used to measure the n_2 of DMA dissolved in chloroform. The wave lengths 457, 488 and 514 nm were used for different polarized lights (i.e. linearly, circularly and elliptically). A quartz cuvette of 2mm width was used for the solution of the sample. A motorized translational stage was used to move the sample from one side of the focal point of the lens to the other smoothly.

III. Results And Discussion

The polarization changeable closed aperture Z-scan Normalized transmittance curve for different polarization of the incident light states at λ=514 nm for the experimental data for the solution of concentration 0.25 is shown in Fig.3. In Fig.4 shows the polarization changeable closed aperture Z-scan Normalized transmittance curve with theoretical fitting for different polarizations of the incident light states at λ=488 nm for the experimental data for the solution of concentration 0.25. Equation (3) is used to obtain the fitted curves by the variation of the value of the nonlinear phase shift $\Delta\varphi_0$. The nonlinear refractive index n_2 were calculated to be the order of ~10^{-6}cm^2/W from the observed transmittance profiles.

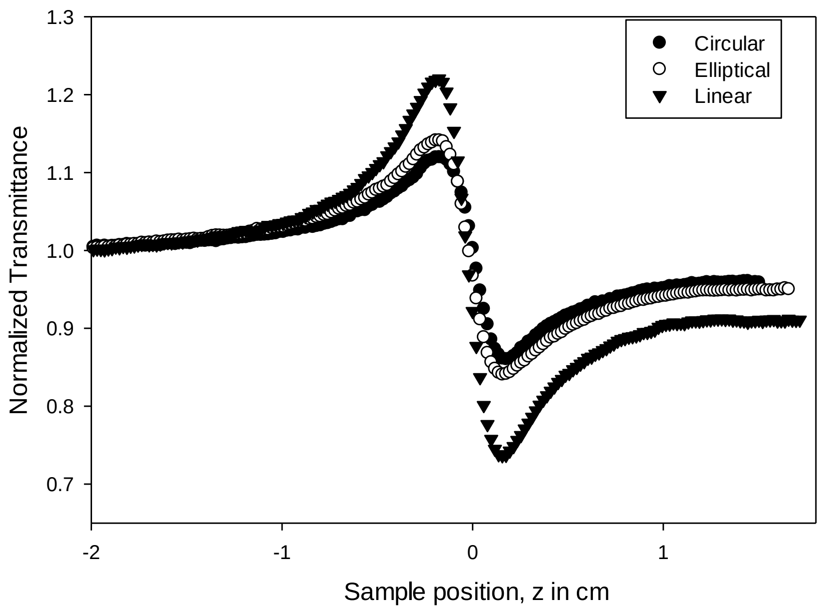

Fig. 3: Normalized transmittance curve of 2,5-dimethylaniline (DMA) for different polarization of the incident light states at λ=488 nm, for the solution of concentration 0.25.

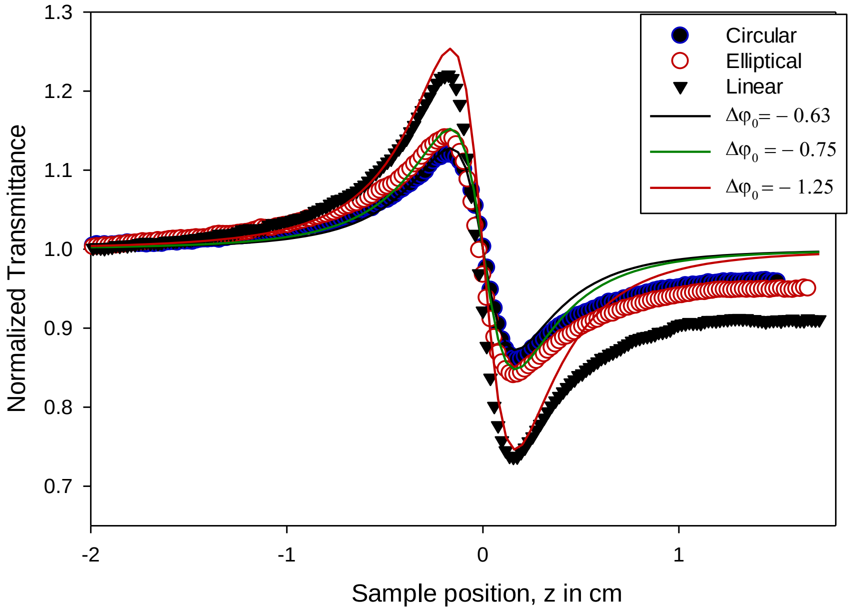

Fig. 4: Normalized transmittance curve with theoritical fitting of 2,5-dimethylaniline (DMA) for different polarization of the incident light states at λ=488 nm, for the solution of concentration 0.25

The Nonlinear phase shift $|\Delta\varphi_0|$ versus intensity I_0 graph for different polarization of the incident light states at λ=514 nm for the solution of concentration 0.33 is shown in Fig.5. The slope was found to be different for the different polarized lights . The nonlinear phase shift was found to vary with different polarized light for all the concentrations of the sample.

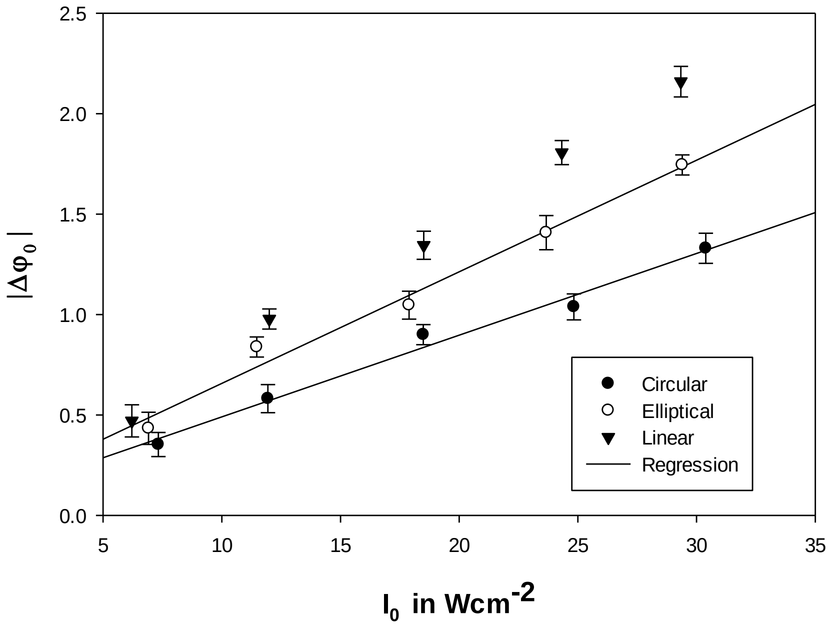

Fig 5:Nonlinear phase shift $|\Delta\varphi_0|$ versus intensity I_0 graph with different polarization of the incident light states at λ=514 nm for the solution of concentration 0.33.

From the observed transmittance profiles the nonlinear refractive indices n_2 , were calculated to be the order of ~10^{-6} cm^2/W as given in Table 1.

Table 1: Using different polarization of the incident lightstates of nonlinear refractive index, n_2 for 2,5-Dimethylaniline (DMA) dissolved in Chloroform at three wavelengths .

Conc. (V/V)	Wavelength, λ (nm)	n_2 (× 10^{-6} cm^2/W)		
		Linear	Elliptical	Circular
0.25	457	-3.57	-3.26	-3.09
	488	-3.39	-3.13	-2.87
	514	-3.13	-2.92	-2.47
0.33	457	-4.55	-4.38	-4.10
	488	-4.43	-3.41	-3.20
	514	-4.28	-3.75	-3.40

It is seen that the normalized transmittance versus sample position plots show that the curve peak is followed by a valley i.e. the nonlinear on-axis phase shift $\Delta \varphi_0$ are negative. Thus, the sign of the nonlinearity is negative for the sample, for the different incident powers of the three wavelengths with different polarized light. The same behavior was observed for the other solutions of the sample.

IV. Conclusion

The nonlinear refractive index n_2 was found to vary with the wavelengths, different polarizations of the incident light states and concentrations of the solution. The response for the nonlinear absorption (measured by the open aperture Z-scan technique) was not found for these concentrations for the existing available powers of the laser [19] .

Before taking the data for the sample 2,5-Dimethylaniline (DMA) in chloroform, the Z-scan data was taken for only chloroform. No nonlinear response was observed for the used experimental powers of the laser. Therefore the value of the nonlinearity found for the sample DMA in chloroform is only for the nonlinear response of DMA.

The normalized transmittance curves of 2,5-dimethyleaniline (DMA) dissolved in chloroform (conc. 0.25) for different polarization states of the incident light at wavelength λ = 488 nm is shown in Fig. 3. Fig.4shows the closed aperture Z-scan transmittance traces with theoretical fitting for different polarizations of the incident light at 488 wavelength for the solution of 0.25 concentration. The nonlinear phase shift $\Delta\varphi_0$ increases with the irradiance on the sample, as shown in Fig. 5. This figure shows that the nonlinear phase shift $\Delta\varphi_0$ for linear polarized light is higher than that for elliptical and circular polarized light. Moreover, the nonlinear phase shift $\Delta\varphi_0$ for elliptical polarized light is higher than that for circular polarized light. When the nonlinear phase shifts are compared, it is the highest for linearly polarized light. Successive measurements with elliptical and circularly polarized light shows a decreasing trend.

The slopes of the plots are used to calculate the nonlinear refractive index. The nonlinear refractive index is highest for linearly polarized light and smallest for circularly polarized light. This is also found for the wavelengths of 457 nm and 514 nm for the same concentration (conc. 0.25) and all the wavelengths of other concentrations (conc. 0.33). The values of the nonlinear refractive indices n_2 are shown in Table 1. The nonlinear refractive indices are found to be of the order ~10^{-6} cm^2/W for all the wavelengths. Though the order of magnitude of the nonlinear refractive index is found to be the same for all concentrations, it shows an increase with increasing concentration of DMA in chloroform, at all the wavelengths for different polarized light states. This is expected from the definition of the susceptibility or polarization [21].

For the nonlinear refraction originating from nonlinear polarization, the nonlinear refractive index is a function of the polarization state of the incident beam [16]. A femto second laser has to be used for probing the nonlinearity arising due to the electronic contribution. But in this paper a continous wave Ar-ion laser has been usedwith the thermal effect being investigated, using linear, elliptical and cicularl polarized light.

Acknowledgments

The authors would like to thank the Higher Education Quality Enhancement project of UGC, Bangladesh for supporting the funds to carry out this research. The cooperation of the department of Chemistry at SUST is highly appreciated. We are also indebted to Prof. M Z Iqbal for useful discussion regarding the experimental data and their analyses.

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Nujhat Nuri Sultana, et al. "Investigation Of Nonlinear Refractive Index For 2,5-Dimethaylaniline (DMA) Under Different Polarized Light." *IOSR Journal of Applied Physics (IOSR-JAP)*, 12(2), 2020, pp. 20-25.