Physical origin of observed nonlinearities in Poly (1-naphthyl methacrylate): Using a single transistor–transistor logic modulated laser beam

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A thermal lens technique is adopted using a single modulated continuous wave (cw) 532-nm laser beam to evaluate the nonlinear refractive index n_2, and the thermo-optic coefficient $\mathrm{d}n/\mathrm{d}T$, in polymer Poly (1-naphthyl methacrylate) (P-1-NM) dissolved in chloroform, tetrahydrofuran (THF), and dimethyl sulfoxide (DMSO) solvents. The results are compared with Z-scan and diffraction ring techniques. The comparison reveals the effectiveness and the simplicity of the TTL modulation technique. The physical origin is discussed for the obtained results.

Keywords: nonlinear refractive index, thermo-optic coefficient, self-phase modulation, nonlinear optics

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1. Introduction

It is well recognized that measurements of thermophysical properties are important for the researchers interested in the optical materials area. The interaction between lasers and matter can result in thermal loading causing degradation and/or reduction of the thermal performance. The changes in properties such as thermal conductivity, thermal diffusivity, and temperature coefficient of optical length with the sample temperature rise define the figure of merit for a given material. The measurements of these properties as a function of temperature with traditional methods are always a challenging goal since it requires high cost devices and an appropriate excitation regime to obtain the data, especially when performed in the temperature interval where the material is submitted to a phase modification.[1,2]

The thermal lens (TL) spectroscopy is one of the most sensitive absorbance methods used in the measurement of a temperature rise following the conversion of the absorbed optical radiation into heat through non-radiative relaxation process.[1] Absorption of a laser beam generates thermal energy in the medium through non-radiative de-excitation resulting in an increase of the temperature of the irradiated region. The temperature distribution of the irradiated region will be the same as the intensity distribution across the beam cross section which is usually Gaussian. Since most liquids have a positive coefficient of thermal expansion, the thermo-optic coefficient, $\mathrm{d}n/\mathrm{d}T$, is negative and consequently the thermal lens generated is a divergent one. The formation of the thermal lens can be probed either by the same pumping beam[3] or by a dual beam.[4] The thermal lens generated by the refractive index when a sample is placed beyond the beam waist ω_0, by one confocal length, $\pi\omega_0^2/\lambda$, is detected by its effect on the intensity at the center of the probe laser beam measured through a pinhole aperture with a photodiode, where λ is the light wave length.

Thermal lens spectroscopy has drawn attention due to its application in measurements of the thermo-optical properties in an optical material. In addition, it was used in two different techniques, the mode matched thermal lens[5,6] and the mode mismatched thermal lens spectropies.[7] It was used in a variety of applications such as scanning microscopy,[8] food analysis, and environmental research,[9,10] realization of optical logical gates,[11] measurements in microchips,[12] microchemical chips,[13] complex fluids,[14] investigation of the quantum yield,[15,16] ion chromatography,[17] and determination of iodine.[18]

Changes in refractive index induced by an optical field can give rise to various nonlinear phenomena in optical materials.[19] In the spatial dimension, the interplay between the divergence of the propagating beam and the nonlinear optical response of the medium can give rise to a wide range of self-action ranging from optical self-trapping to spontaneous formation of ring patterns due to the modulation instability.[20] A related phenomenon is the spatial self-modulation of a coherent beam which generates concentric intensity rings in the far field. The diffraction rings have been observed in nematic liquid crystals by Zolotko *et al.*,[21] in thermally dependent refractive index,[22] polymer films,[23] in Kerr media[24,25] and liquid crystals.[26,27]

The diffraction ring pattern is understood from the spatial self-modulation effect. A pump beam with a Gaussian intensity profile should induce a phase shift $\Delta\Phi$, with a bell-shaped transverse profile. For each point y_1, on the Gaussian distribution of the beam, another point profile y_2 exists, with the

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same slope. The radiation field in the regions around y_1 and y_2 have the same wave vector and should interfere constructively or destructively when $\Delta\Phi(y_1)-\Delta\Phi(y_2)=m\pi$, if m is even or odd respectively.

The nonlinear response of a medium can be characterized using the Z-scan technique developed by Sheik-Bahae *et al.*[28] This technique employs a single Gaussian beam in a tight focus geometry to measure the transmittance of a nonlinear medium as a function of the sample position. The Z-scan technique is based on the self-focusing or defocusing of a converging beam of known spatial structure induced by moving a nonlinear sample along the light propagation direction (z axis). This technique permits a rapid evaluation of the magnitude and sign for both the real (nonlinear refraction, NLR) and imaginary (nonlinear absorption, NLA) parts of the nonlinearity of transparent solids and liquids. As far as NLR is concerned, the transmission across the aperture in the far field is registered as a function of the sample position along the z axis (closed aperture, CA), while NLA can be inferred either by using CA Z-scan or more accurately, by independently measuring the overall sample transmission (open aperture, OA).[29,30] The on-axis phase shift $\Delta\Phi$, the nonlinear refractive index n_2, and nonlinear absorption coefficient β, can be estimated by using this method.

In this paper, the observation of a thermal lens effect is reported, using a cw transistor–transistor logic (TTL) modulated laser beam with 532-nm wavelength in the polymer Poly (1-naphthyl methacrylate) (P-1-NM) solutions. The calculated nonlinear refractive index n_2, and thermo-optic coefficient $\mathrm{d}n/\mathrm{d}T$, are compared with those calculated by using the Z-scan and diffraction ring techniques.

2. Theory

2.1. Thermal lens technique

Consider the transmission of optical radiation with certain intensity through an absorbing medium. In TL technique, the thermal gradient established after absorption and thermal relaxation of the sample results in a change of intensity at the beam center owing to the incident beam divergence. The thermal lens signal is expressed as the relative change in power.[31] A convenient quantity for the experiments is the total fractional intensity change

$$\frac{I(x,t)-I(x,t=\infty)}{I(x,t=\infty)} = \frac{\left\{1-(\theta/2)\tan^{-1}\left[2x/[3+x^2+(9+x^2)(t_\mathrm{c}/2t)]\right]\right\}^2}{\left\{1-(\theta/2)\tan^{-1}\left[2x/(3+x^2)\right]\right\}^2}-1, \quad (1)$$

where $I(x,t)$ is the intensity measured at the beam center, $I(x,t=\infty)$ is the intensity measured at the detector after sufficient time when a steady state temperature difference is reached $x=z/z_0$ and

$$\theta=\frac{P\alpha L_\mathrm{eff}}{\lambda k}\frac{\mathrm{d}n}{\mathrm{d}T}, \quad (2)$$

where z is the distance between the sample and the waist of the beam z_0 is the Rayleigh range, α is the linear absorption coefficient, L_eff is the effective thickness of the sample which is given by ($L_\mathrm{eff}=(1-\mathrm{e}^{-\alpha L})/\alpha$), L is the sample thickness, P is the laser input power, λ is the pump laser wavelength, k is the thermal conductivity, and $\mathrm{d}n/\mathrm{d}T$ is the thermo-optic coefficient.

The characteristic thermal time of the medium t_c, can be written as

$$t_\mathrm{c}=\frac{\omega_0^2}{4D}. \quad (3)$$

Here, ω_0 is the beam radius at the sample and D is the thermal diffusivity which can be written as

$$D=\frac{k}{\rho C}, \quad (4)$$

where ρ is the sample density and C is the specific heat of the sample.

For the thermal nonlinearity and steady state case, the on-axis change in the refractive index Δn, can be expressed as[32]

$$\Delta n=\frac{\mathrm{d}n}{\mathrm{d}T}\frac{I\alpha\omega_0^2}{4k}. \quad (5)$$

In addition, Δn can be related to the total refractive index of the medium n, and the background refractive index n_0 as[33]

$$n=n_0+\Delta n, \quad (6)$$

and

$$\Delta n=n_2 I, \quad (7)$$

where n_2 is the nonlinear refractive index and I is the laser input intensity ($=2p/\pi\omega_0^2$).

2.2. Diffraction ring technique

Due to the average on-axis refractive index change Δn, the beam traversing the medium acquires nonlinear phase-shift $\Delta\Phi$, given by[32]

$$\Delta\Phi=\Delta n k_0 L_\mathrm{eff}, \quad (8)$$

where $k_0(=2\pi/\lambda)$ is the beam wave vector in a vacuum.

The on-axis nonlinear phase-shift $\Delta\Phi$, can be related to the number of rings observed N as[34]

$$\Delta\Phi=2\pi N. \quad (9)$$

Combining Eq. (4) and Eqs. (7)–(9), one can calculate Δn, n_2, and $\mathrm{d}n/\mathrm{d}T$.

2.3. *Z*-Scan technique

From the open aperture (OA) Z-scan technique, the nonlinear absorption coefficient β, can be calculated using the following formula[28]

$$\beta = \frac{2\sqrt{2}\Delta T}{IL_{\text{eff}}}. \tag{10}$$

Here, ΔT is the one-valley transmittance on the (OA) Z-scan curve.

The difference between the normalized peak and the valley transmittance $\Delta T_{\text{p-v}}$, can be written as[28]

$$\Delta T_{\text{p-v}} = 0.406(1-S)^{0.25}\,|\Delta\Phi|\,. \tag{11}$$

and

$$S = 1 - \exp\left(\frac{-2r_a^2}{\omega_a^2}\right). \tag{12}$$

Here, $\Delta\Phi$ is the on-axis phase shift at the focus, S is the linear transmittance of the aperture, r_a is aperture radius, and ω_a is the beam radius at the aperture. The nonlinear refractive index n_2, can be approximated using the following equation:[35]

$$n_2 = \frac{\lambda\Delta\Phi}{2\pi IL_{\text{eff}}}. \tag{13}$$

3. Experiment

3.1. Sample preparation

The poly (1-naphthyl methacrylate) was prepared according to Aspler and Guillet[36,37] by the free radical polymerization of 1-naphthyl methacrylate in a benzene solution degassed to ≤ 0.005 Torr (1 Torr = 1.33322×10^2 Pa) residual pressure. Decanoyl peroxide was used as an initiator. The polymer was polymerized to about 100% yield for 70 h at 70 °C and purified by multiple precipitations from benzene into methanol and then freeze dried from benzene. The average molecular weight was Mn = 132 000. Figure 1 shows the structure of the polymer under the present study.

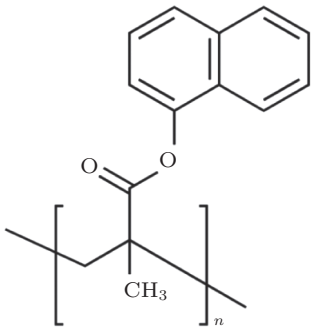


Fig. 1. The chemical structure of Poly (1-naphthyl methacrylate) (P-1-NM).

3.2. Spectroscopic studies

The spectral properties of the P-1-NM in the solvents chloroform, tetrahydrofuran (THF), and dimethyl sulfoxide (DMSO) are studied by recording the absorption spectra of the P-1-NM solutions using Cecil ReflectaScan Reflectance Spectrophotometer CE-3055. The absorption spectra of the P-1-NM in the solvents chloroform, THF and DMSO are shown in Fig. 2. The spectrum of the optical absorption was computed from the absorbance data. The values of absorption coefficient α, at a wavelength of 532 nm for 0.05 mM concentration of P-1-NM in chloroform, THF, and DMSO are 15.81 cm^{-1}, 14.4 cm^{-1}, and 10.05 cm^{-1}, respectively.

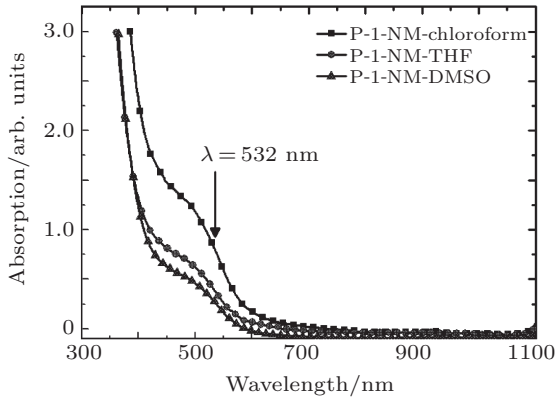


Fig. 2. The absorption spectrum of P-1-NM in the solvents chloroform, THF, and DMSO.

3.3. Experimental setup

The Poly (1-naphthyl methacrylate) was prepared in a glass cell of 1-mm thickness as the nonlinear material in the experiment. The apparatus consists of a diode laser

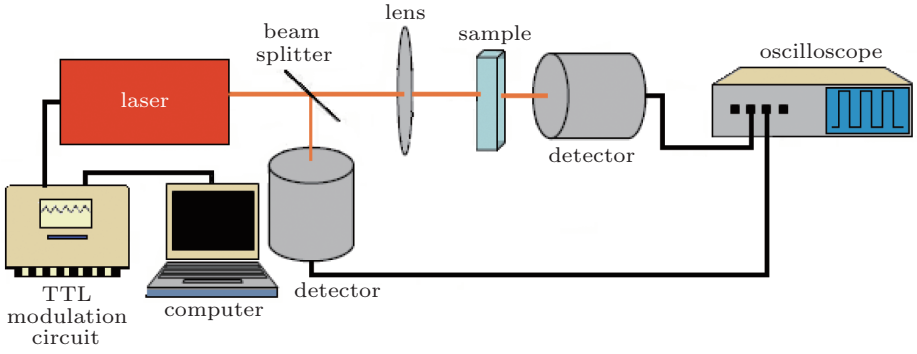


Fig. 3. (color online) Thermal lens experimental setup.

4-mW cw output, beam radius 1.5 mm at $1/e^2$, 532-nm wavelength, a positive glass lens of +50-mm focal length, a glass cell 1-mm thickness filled with P-1-NM solutions, a transparent screen of 30 cm×30 cm, a digital camera, two power meters to measure input and transmitted output beam powers through the cell, a frequency generator model (EM1634) and an oscilloscope model (lodestar-MOS-620CH). For the ring pattern the output power meter is replaced by the screen. Output of the laser is modulated at 20 Hz for the TL measurements. Figure 3 shows the experimental setup.

4. Results

4.1. TL measurements

The transient signals of Poly (1-naphthyl methacrylate) in chloroform, THF, and DMSO are shown in Fig. 4 for a pumping power 4 mW at room temperature. The solid line corresponds to the data fitting of Eq. (1) to the TL experimental one with θ, t_c as adjustable parameters. The values obtained for θ and t_c are given in Table 1. Using Eqs. (3)–(7), the values for D, n_2, $\mathrm{d}n/\mathrm{d}T$, and Δn are calculated and shown in Table 1.

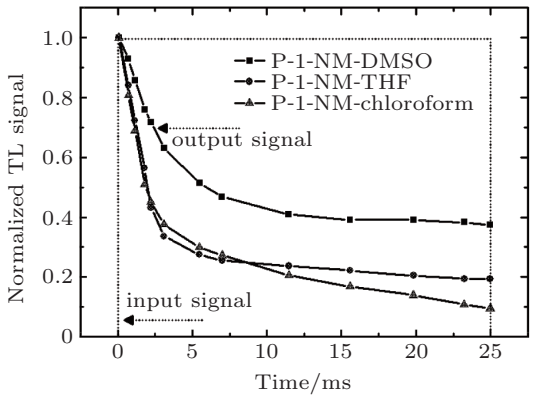


Fig. 4. Transient signals of (P-1-NM) solutions, dotted curve represents the input signal.

Table 1. Characteristic TL time constant (t_c), thermal conductivity (K), thermally induced phase shift (θ), thermal diffusivity (D), nonlinear refractive index (n_2), and thermo-optic coefficient ($\mathrm{d}n/\mathrm{d}T$) are obtained for (P-1-NM) solvents.

*Solvent	k/W·m^{-1}·K^{-1}	t_c	θ/rad	$D/10^{-4}$ cm^2·s^{-1}	$n_2/10^{-7}$ cm^2·W^{-1}	$(\mathrm{d}n/\mathrm{d}T)/10^{-5}$ K^{-1}	$\Delta n/10^{-4}$
Chloroform	0.117	3.43	2.741	3.41	8.49	5.37	0.046
THF	0.126	2.252	2.435	5.19	7.15	5.34	0.038
DMSO	0.199	9.416	2.155	1.25	5.31	8.99	0.028

*The standard error of all estimated optical properties was ±1.165 %, ±3.103%, and ±4.986% for Chloroform, THF, and DMSO, respectively.

4.2. Diffraction ring pattern measurements

The experimental setup for the diffraction ring patterns is the same as that previously mentioned, except replacing the output power meter by the semi-transparent screen. The on-axis index change Δn has been calculated using the relation[22]

$$\Delta n = \frac{\Delta \Phi}{k L_{\mathrm{eff}}}. \tag{14}$$

As mentioned in Table 2, the number of rings, N, observed are 7, 5, and 2, for the P-1-NM solutions respectively. Based on N, the parameters $\Delta\Phi$, Δn, n_2, and $\mathrm{d}n/\mathrm{d}T$ are calculated and the values obtained are given in Table 2. The diffraction ring patterns for the P-1-NM solutions are shown in Fig. 5.

Table 2. Nonlinear optical parameters for (P-1-NM) solutions determined by using the diffraction ring technique.

Solvent	N	$\Delta\Phi$	Δn	n_2/cm^2·W^{-1}	$(\mathrm{d}n/\mathrm{d}T)$/K^{-1}
Chloroform	7	0.76	1.29×10^{-4}	2.37×10^{-7}	1.50×10^{-5}
THF	5	0.54	0.876×10^{-4}	1.61×10^{-7}	1.20×10^{-5}
DMSO	2	0.21	0.29×10^{-4}	0.54×10^{-7}	0.91×10^{-5}



(a) (b) (c)

Fig. 5. (color online) Diffraction ring patterns for the P-1-NM solutions. (a) Chloroform. (b) THF. (c) DMSO.

4.3. Z-Scan measurements

The Z-scan experiments were performed using the same laser (532 nm) which was focused by the same lens (+50 mm). The laser beam waist ω_0, at the sample is measured to be 21.63 μm and the Rayleigh length z_0, is 2.76 mm, aperture radius r_a, is 2 mm, the radius of the beam spot at the aperture ω_a, is 3.82 mm, the distance from the focal point to the detector z_a is 250 mm, input power p, is 4 mW which lead to an input intensity I, of 544.05 W/cm^2. The linear transmittance at the aperture S, is 0.42.

Figure 6(a) shows the normalized transmittance of P-1-NM solutions, which was measured using the open aperture Z-scan technique. The sample exhibited a reduction in the transmission near the focus of the lens which is indicative of a nonlinear absorption. Closed aperture Z-scan transmittance curves of the samples are shown in Fig. 6(b). The peak-to-valley structure of the closed aperture Z-scan curve in Fig. 6(b) implies that the sample possesses a negative nonlinear refraction property (self-defocusing).

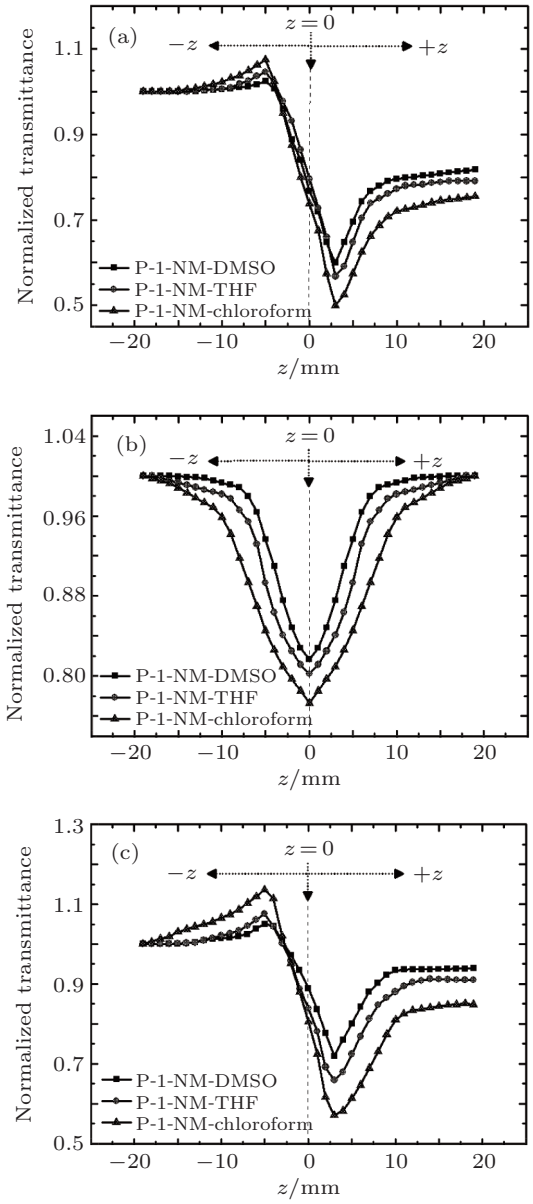


Fig. 6. Z-scan experimental data for P-1-NM solutions: (a) closed aperture (CA), (b) open aperture (OA), and (c) CA/OA .

If a nonlinear absorption is present, as in our case by dividing the curves obtained from the two Z-scan modes (closed aperture/open aperture) one can extract the pure nonlinear coefficient β, thermo-optic coefficient, $\mathrm{d}n/\mathrm{d}T$ and nonlinear refractive index n_2. These parameters are calculated from the open and closed aperture normalized transmittances in Figs. 6(a) and 6(c) respectively, and are given in Table 3.

Table 3. Nonlinear optical parameters for (P-1-NM) solutions determined by Z-scan technique.

Solvent	$\Delta T_{\mathrm{p-v}}$	L_{eff}/cm	$\Delta\Phi$	n_2 /10^{-7} cm^2·W^{-1}	$\Delta n/10^{-4}$	$(\mathrm{d}n/\mathrm{d}T)$ /10^{-5} K^{-1}
Chloroform	0.565	0.050	1.59	4.94	2.68	3.12
THF	0.416	0.053	1.17	3.44	1.87	2.57
DMSO	0.330	0.063	0.93	2.29	1.25	3.88

5. Discussion

The physical and chemical properties of organic molecules used in different scientific applications can strongly depend on the properties of the surrounding media.[38,39] For liquid solutions, the solvent plays a fundamental role in the photo-physical processes, leading to the modification of the ground and excited-state energies of the molecules. The interaction of solute with the surrounding solvent leads to the reorientation of the solvent molecules around a solute molecule. The result of solute interaction with solvent molecules depends on the nature of arising forces (as hydrogen bonding in specific or universal interaction in general) which are determined by charge distribution and polarizability of the solvent and solute molecules. Considerable changes in the energy of the solvated solute molecules may occur with changes in the solvent, especially in polar media. The values of absorption at $\lambda = 532$ nm of Poly (1-naphthyl methacrylate) (P-1-NM) are derived from the absorption bands in chloroform, THF, and DMSO that are 0.774, 0.654, and 0.404 respectively. According to the absorption values the maximum change in P-1-NM solution temperature should occur with chloroform, then it decreases towards DMSO. Accordingly, the number of rings N, decreases which leads to a reduction in the change of refractive index Δn, hence in the nonlinear refractive index n_2, and in the thermo-optic coefficient $\mathrm{d}n/\mathrm{d}T$. The obtained results agree well with the trend of results achieved by the three different techniques suggested in this work, although the specific results for the three parameters, i.e., Δn, n_2, and $\mathrm{d}n/\mathrm{d}T$ calculated show some discrepancy.

6. Physical origin of observed nonlinearities

The thermal properties of P-1-NM solutions are mainly governed by the solvent. Therefore, the specific heat C, the density ρ, and the thermo-optic coefficient $\mathrm{d}n/\mathrm{d}T$ are all properties which are characteristics of the solvent. Thus, assuming that a change in temperature of the dye solution primarily leads to a change in the density of solution, so the variation of $\mathrm{d}n/\mathrm{d}T$ for various solvents can be written as

$$\frac{\mathrm{d}n}{\mathrm{d}T} = \frac{\delta n}{\delta \rho}\frac{\delta \rho}{\delta T} = -\Gamma\rho\left(\frac{\delta n}{\delta \rho}\right), \tag{15}$$

where Γ is the expansivity. The change in refractive index with density can be determined from Lorentz–Lorenz relationship.[40] The increase of absorption coefficient leads to an increase of the absorbed amount of laser power by the solution and as a result heat produced increases monotonically. This will enhance the thermal nonlinear refractive index. As the thermal conductivity decreases, it leads to a local accumulation of resultant heat via the absorption, hence the enhancement of thermal effects. It is known that thermal nonlinear-

ity can be reduced via the use of solvents with high specific heats.[41]

According to the results obtained from the three methods used in this work on P-1-NM in three species of solvents, it was found that the nonlinear refractive index n_2, and the thermo-optic coefficient $\mathrm{d}n/\mathrm{d}T$, of P-1-NM solutions are dependent on the surrounding medium parameters, namely polarity, dipole moment, thermal conductivity, specific heat, and absorption coefficient, as shown in Table 4.

The thermal contributions of the electric field must be considered. This effect contributes to a negative nonlinear refractive index. As the electric field is absorbed by the medium, accumulative thermal heating causes a decrease in the index of refraction. This process is a fluence dependent on the refractive index change with time. It is also possible for an acoustic wave to be generated by the nonlinear absorption in the medium. This wave spreads the beam waist radius and again causes a decrease in the refractive index.

Table 4. The important physical properties of the solvents used in the present work (1 deb=10^{-18} Fr·cm).

Solvent	Polarity index	Dipole moment/deb	Density /gm·L^{-1}	Thermal conductivity k/W·m^{-1}·K^{-1}	Specific heat C/J·g^{-1}·K^{-1}	Absorption coefficient α/cm^{-1}
DMSO	7.2	3.96	1.092	0.199	1.958	10.05
THF	4	1.75	0.888	0.126	1.058	14.4
Chloroform	4.4	1.040	1.492	0.117	0.9566	15.81

7. Conclusion

Thermal lens, cw TTL modulated laser beam, experiments are carried out in a polymer Poly (1-naphthyl methacrylate) (P-1-NM) in two species of polar aprotic solvents (DMSO and THF) and non-polar solvent (Chloroform). The nonlinear refractive index n_2, and thermo-optic coefficients $\mathrm{d}n/\mathrm{d}T$, obtained are large and negative in nature and are strongly dependent on the properties of the solvent such as polarity, dipole moment, thermal conductivity, specific heat, and the absorption coefficient. The nonlinear refractive indices and thermo-optic coefficients of the three solutions are found to be of the order of 10^{-7} cm^2/W and 10^{-5} K^{-1} respectively. These results compared well with those obtained by the Z-scan and diffraction rings techniques. Based on the experimental findings, it appears that chloroform is the best candidate used for nonlinear studies.

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