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Theoretical Estimation of Third Order Optical Nonlinearity of poly (1-Naphthyl Methacrylate)

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ABSTRACT

Diffraction ring technique has been employed to estimate the nonlinear refractive index (n_2), the second order nonlinear index of refraction ($\bar{n}_2$) and the third order nonlinear optical susceptibility ($\chi^{(3)}$) for poly (1-naphthyl methacrylate) (P-1 NM) dissolved in chloroform, tetrahydrofuran (THF) and dimethylsulfoxide (DMSO) solvents. The technique estimates the value of n^2 and $\chi^{(3)}$ for P-1 NM in chloroform, THF and DMSO is of the order of 10^{-2} m²/W, 10^{-4} Fm²/Ws and 10^{-4} m²/V² respectively.

Keywords: Third Order Optical Susceptibility, Nonlinear Refractive Index, Second Order Index of Refraction, Diffraction Ring Technique.

I. INTRODUCTION

Materials which possess third order nonlinear optical characteristics are vital because they have potential applications in optical information processing, optical sensor manufacturing applications, optical computing and data storing [1-2]. These nonlinear materials are drawing attentions in research fields. The nonlinear refractive index (n_2) and the second order nonlinear index of refraction ($\bar{n}_2$) are two optical parameters that characterize the third order nonlinear optical susceptibility $\chi^{(3)}$ [3-7].

Passage of intense optical beam through third order nonlinear optical medium generates various phenomena that can be used to characterize third order susceptibility $\chi^{(3)}$. When a very strong laser beam is passed through such medium, interaction of the beam with the medium leads to the spatial phase modulation of the beam. This phase modulation is commonly known as 'self-phase modulation' because the beam itself controls its own phase and propagation. Consequence of the self-phase modulation is to produce concentric rings in far field region of the beam which is termed as diffraction ring [8-10]. Numbers of diffraction ring is extensively related to the nonlinear refractive index and hence other optical constants can be estimated using diffraction ring technique. Formation of diffraction ring is a common phenomena that has been observed in various liquid crystals, organic liquids, Kerr media and polymer films by various researchers [10-12].

In this work nonlinear refractive index (n_2), second order index of refraction ($\bar{n}_2$) and third order nonlinear optical susceptibility ($\chi^{(3)}$) has been calculated theoretically using diffraction ring technique for poly (1-naphthyl methacrylate). The result has been estimated for dissolved sample in chloroform, tetrahydrofuran (THF) and dimethylsulfoxide (DMSO). A continuous wave laser capable of delivering 4mW optical power at 532 nm has been used to perform the calculation.

II. THEORY

Laser beam with transverse Gaussian intensity profile can be described by the following equation

$$E(\rho,z)=E_0\frac{W_0}{W(z)}\exp\left[-\frac{\rho^2}{W^2(z)}\right]\exp\left[-jk\left(z+\frac{\rho^2}{2R(z)}-\frac{\xi(z)}{k}\right)\right].......(1)$$

Where E_0 and W_0 are beam amplitude and beam waist respectively. ρ is called radial coordinate for any transverse plane described by $\rho = (x^2 + y^2)^{1/2}$, wave number k = 2π/λ where λ is the laser wavelength, z is the beam propagation direction, R(z) is the radius of curvature of the wave front of the laser beam and ξ(z) is the phase retardation relative to the plane wave [10, 11]. The beam intensity is function of the axial distance (z) and radial coordinate (ρ). Output electric field due to the interaction of the beam with sample is given by,

$$E_0 = E(r,z)\exp\left(-\frac{\alpha L}{2}\right)\exp[-j\Delta\varphi(\rho)].......(2)$$

Where α is the linear absorption coefficient of the sample, L is the geometrical path length of the sample and Δφ is the nonlinear phase change induced by the nonlinear medium. Equation (2) shows decrease of the output electric field due to the linear absorption of the medium. The nonlinear phase change Δφ is related to the on-axis maximum phase change $\Delta\varphi_0$ by the following equation,

$$\Delta\varphi(\rho)=\Delta\varphi_0\exp\left[-\frac{\rho^2}{W^2(z)/m}\right].......(3)$$

m is a parameter used to characterize the local or nonlocal nonlinear optical response [10, 11].

The On-axis phase change Δφ depends on the axial distance z given by the following equation [11].

$$\Delta\varphi(z,m)=\frac{\Delta\Phi}{\left[1+(z/z_0)^2\right]^{m/2}}.......(4)$$

Phase change (ΔΦ) at z = 0 is given by,

$$\Delta\Phi = k_0\Delta n(0,0)L_{eff}.......(5)$$

Here k_0 is the beam wave vector in vacuum, Δn(0,0) is the on-axis refractive index change at z = 0, L_{eff} is the effective length given by the equation,

$$L_{eff}=\frac{1-e^{-\alpha L}}{\alpha}.......(6)$$

The on-axis maximum nonlinear phase change (at z = 0) is associated with the number of diffraction rings (N) formed in the far field region [13-15],

$$\Delta\Phi = 2\pi N.......(7)$$

Thus the nonlinear refractive index n_2 can be estimated from the following relationship,

$$n_2=\frac{\lambda\Delta\Phi}{2\pi L_{eff}}........(8)$$

Here n_0 is the usual or low intensity refractive index, ϵ_0 and c are the permittivity of free space and light velocity in vacuum respectively [16-17].

Furthermore the third order nonlinear optical susceptibility $\chi^{(3)}$ has been calculated by the following relationship.

$$n_2=\frac{3\chi^{(3)}}{4n_0^{\ 2}\varepsilon_0 c}.................(10)$$

III. DIFFRACTION RING TECHNIQUE

Diffraction ring technique experiment requires a biconvex lens which directs the Gaussian beam to be focused on the sample. The sample is allowed to move along the beam propagation direction, about the focal point of the lens, so that the sample is set in the convergent or divergent beam region. The concentric rings are formed at the far field region as a result of the beam self phase modulation.

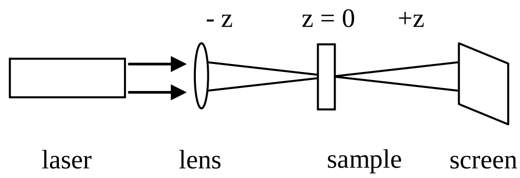

Figure 1. Experimental setup to observe diffraction rings.

IV. CALCULATION

Table 1. Estimated values of n_2, $\overline{n}_2$ and $\chi^{(3)}$ of poly (1-naphthyl methacrylate) (P-1 NM) dissolved in chloroform.

On axis Refractive Index Change ($\Delta n \times 10^{-4}$)	Nonlinear Refractive Index ($n_2 \times 10^{-2}$) m^2/W	Second Order Index of Refraction ($\overline{n}_2 \times 10^{-4}$) Fm^2/Ws	Third Order Susceptibility ($\chi^{(3)} \times 10^{-4}$) m^2/V^2
0.37	0.93	0.51	1.12
0.74	1.85	1.03	2.25
1.12	2.79	1.54	3.37
1.49	3.72	2.05	4.49
1.86	4.65	2.57	5.62

Table 2. Estimated values of n_2, $\overline{n}_2$ and $\chi^{(3)}$ of poly (1-naphthyl methacrylate) (P-1 NM) dissolved in tetrahydrofuran (THF).

On axis Refractive Index Change ($\Delta n \times 10^{-4}$)	Nonlinear Refractive Index ($n_2 \times 10^{-2}$) m^2/W	Second Order Index of Refraction ($\overline{n}_2 \times 10^{-4}$) Fm^2/Ws	Third Order Susceptibility ($\chi^{(3)} \times 10^{-4}$) m^2/V^2
0.36	0.88	0.49	1.07
0.71	1.77	0.98	2.15
1.07	2.67	1.47	3.22
1.42	3.55	1.96	4.29
1.78	4.44	2.45	5.37

Table 3. Estimated values of n_2, $\overline{n}_2$ and $\chi^{(3)}$ of poly (1-naphthyl methacrylate) (P-1 NM) dissolved in dimethylsulfoxide (DMSO).

On axis Refractive Index Change ($\Delta n \times 10^{-4}$)	Nonlinear Refractive Index ($n_2 \times 10^{-2}$) m^2/W	Second Order Index of Refraction ($\overline{n}_2 \times 10^{-4}$) Fm^2/Ws	Third Order Nonlinear Optical Susceptibility $\chi^{(3)} \times 10^{-4}$ m^2/V^2
0.15	0.38	0.21	0.46
0.45	1.13	0.63	1.37
0.76	1.89	1.04	2.28
1.06	2.65	1.46	3.20
1.36	3.41	1.88	4.11

V. GRAPHS

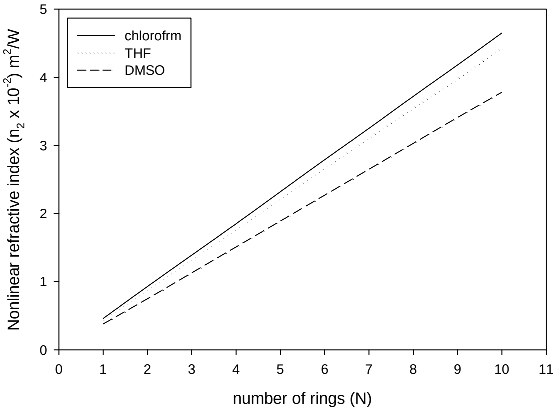

Figure 2. Variation of nonlinear refractive index as function of diffraction ring numbers.

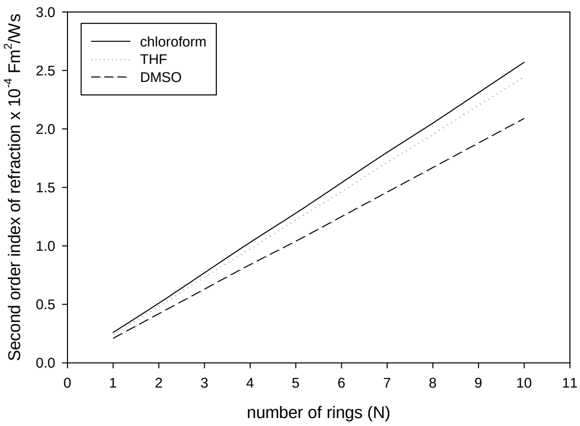

Figure 3. Variation of second order index of refraction for various numbers of diffraction ring.

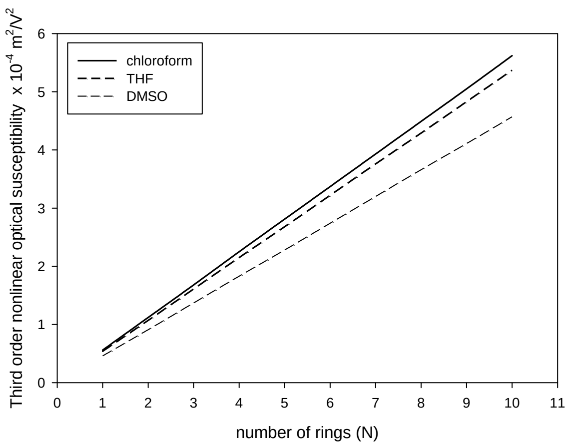

Figure 4. Third order nonlinear optical susceptibility for various numbers of diffraction ring.

VI. RESULT

The diffraction ring technique provides the estimated value of the nonlinear refractive index (n_2) of poly (1-naphthyl methacrylate) (P-1 NM) dissolved in chloroform, THF and DMSO of the order of 10^{-2} m^2/W. Order of the estimated value of n_2 of (P-1 NM) indicates that the origin of the optical nonlinearity is thermal [16]. In addition, the calculated second order index of refraction($\bar{n}_2$) and third order nonlinear optical susceptibility ($\chi^{(3)}$) of P-1 NM in chloroform, THF and DMSO was found to be of the order of 10^{-4} Fm^2/Ws and 10^{-4} m^2/V^2 respectively. The values of n_2, $\bar{n}_2$ and $\chi^{(3)}$ increases as the number of diffraction ring increases for the case of all solvents. Estimated values of the nonlinear phase shift ($\Delta\varphi$) and on axis nonlinear phase change (Δn) for P-1 NM dissolved in three different solvents has been presented in table 1, 2 and 3.

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