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We report the nonlinear absorption and refraction of banana shaped liquid crystal, namely 1,3-phynelyne bis [N-(2-hydroxy-4-n-decyloxy-benzylidene)-4'-aminobenzoate] using Z-scan technique. The measurements were carried out using a Q-switched, frequency doubled Nd:YAG laser producing 7 nanosecond laser pulses at 532 nm. The Z-scan result shows that the compound exhibits negative non-linearity. The nonlinear refractive index and nonlinear absorption coefficient of this compound were found to be -1.283×10^{-11} esu and 2.232 cm/GW, respectively. The real and imaginary parts of the third-order nonlinear optical susceptibility were of the order of 10^{-13} esu. The results suggest that these molecules are potential nonlinear optical materials for device applications.

Keywords Banana shaped liquid crystal; nonlinear optical property; Q-switching; Z-scan

1. Introduction

Materials with good nonlinear response are interesting for wide range of photonic applications, such as optical limiting, optical switching etc. Syntheses of new nonlinear optical materials have therefore attracted the interest of several research groups. Liquid crystals occupy an important niche in nonlinear optics as result of their unique physical and optical properties. Liquid crystals are known to exhibit large optical nonlinearities which have been studied extensively, because of their potential applications to photonics, such as high speed optical switching devices, real time coherent optical signal processors, optical storage, optical limiting and photorefraction [1,2]. Liquid crystals are very attractive for applications in tunable optical devices, since they demonstrate nonlinear optical behavior arising from molecular reorientation and/or thermal effects [3,4]. Since liquid crystals molecules typically

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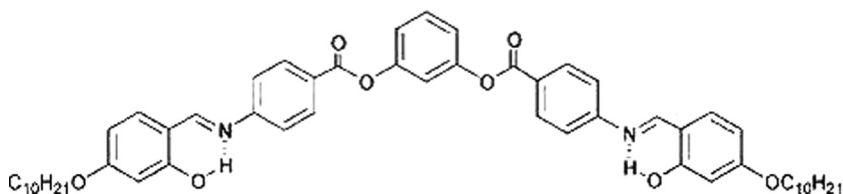
Figure 1. Structure of liquid crystal.

have delocalized π-electrons, they are expected to be potential sources of fast and large nonlinearities [5].

In this paper we present an investigation of the third-order nonlinear optical properties, in the B_2 phase of banana shaped liquid crystal, 1,3-phynelyne bis [N-(2-hydroxy-4-n-decyloxy-benzylidene)-4′-aminobenzoate] by single beam Z-scan technique. The chemical structure is as shown in Figure 1. Prof. C. V. Yalemaggad of CLCR, Bangalore, India, supplied this sample. Differential scanning calorimetry (DSC) and X-ray and electrical studies have been carried out by Yalemaggad *et al.* [8].

2. Experiment

The linear absorption spectrum of the banana shaped liquid crystal was obtained at room temperature by using the UV–Visible fiber optic spectrometer (Model SD2000, Ocean Optics Inc.), Single beam Z-scan technique [6], was employed to measure the third-order optical nonlinearities of the liquid crystal sample. This technique enables simultaneous measurement of nonlinear refraction (NLR) and nonlinear absorption (NLA). Basically, in this technique a Gaussian laser beam is focused, using a lens, on the cuvette containing the liquid sample. The cuvette is translated across the focal region and changes in the far-field intensity pattern are monitored. The experiments were performed using a Q-switched, frequency doubled Nd: YAG laser (Spectra-Physics GCR170) which produces 7 ns pulses at 532 nm and at a pulse repetition rate of 10 Hz. Using a lens of 25 cm focal length focused the laser beam. The laser beam waist at the focused spot was estimated to be 18.9 μm and the corresponding Rayleigh length is 2.11 mm. The Z-scan measurements were carried out using a cuvette of 1 mm thickness, which is less than the Rayleigh length. Hence, the thin sample approximation is valid. The Z-scan experiment was performed at an input peak-intensity of 2.39 GW/cm^2. The nonlinear transmission of the liquid crystal sample, with and without the aperture in front of the detector was measured in the far-field using Laser Probe Rj-7620 Energy Meter with Pyroelectric detectors. For the Z-scan experiments the liquid crystal sample was dissolved in Dimethylformamide (DMF) and solution of concentration $1 \cdot 10^{-3}$ mol/L was used.

3. Results and Discussions

Figure 2 shows the UV-Visible spectrum of the banana shaped liquid crystal, which clearly shows negligible single photon absorption at 532 nm wavelength. Therefore, the nonlinear optical measurements carried out in our experiments are under non-resonant electronic excitation of the molecules.
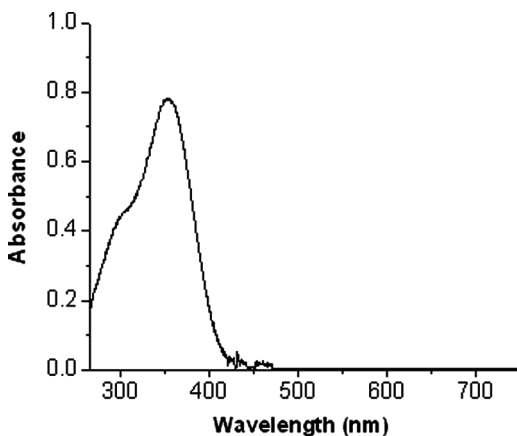
Figure 2. UV-Visible absorption spectrum of the banana shaped liquid crystal.

The nonlinear transmission of compounds without aperture (open aperture) was measured in the far field as the sample was moved through the focal point. This allows us to determine the nonlinear absorption β. The open aperture curve of liquid crystal sample is shown in Figure 3(a). Here, the transmission is symmetric with respect to focus (z = 0), where it has a minimum transmission, showing an intensity dependent absorption effect. The shape of the open aperture curve suggests that the compound exhibits two-photon absorption [7,9,15].
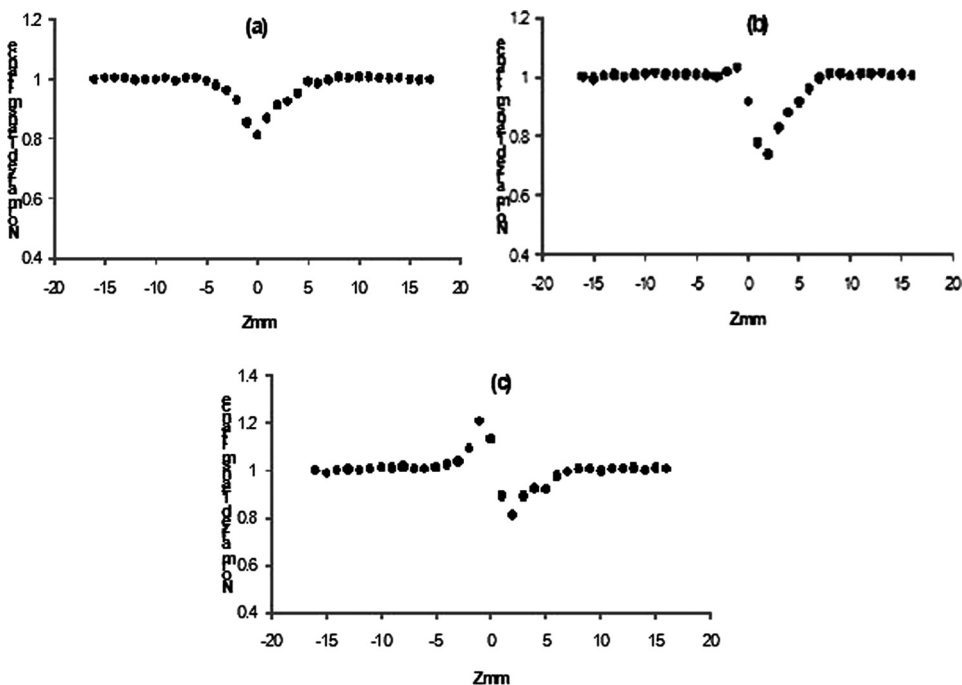

Figure 3. (a) Open aperture, (b) Closed aperture and (c) Pure nonlinear refraction Z-scan curves of banana shaped liquid crystal.

The normalized transmittance for the open aperture Z-scan is given by [6].

$$T(z) = \frac{\ln[1 + q_0(z)]}{q_0(z)} \quad \text{for } |q_0(z)| < 1 \tag{1}$$

Where,

$$q_0(z) = \frac{I_0 \beta_{\text{eff}} L_{\text{eff}}}{(1 + z^2/z_0^2)} \tag{2}$$

and α is the linear absorption coefficient, L is the thickness of the sample, I_0 is the on axis peak irradiance at the focus, and z_0 is the Rayleigh length.

It is known that nonlinear absorption coefficient β, depends on the number of absorptive centers in a unit volume. Assuming that this number is N_0, in units of cm^{-3}, we have [10,11],

$$\beta = \sigma_2 N_0 = \sigma_2 N_A d \times 10^{-3} \tag{3}$$

Here, N_0 is the molecular density of the sample (in units of 1/cm^3), σ_2 is the molecular TPA coefficient (or cross-section) (in units of cm^4/GW), d is the concentration of the liquid crystal sample and N_A is the Avogadro number. For known β and d, the value of σ_2 can be obtained. Molecular TPA cross-section (in units of cm^4 s/photon) is also defined as [11,12],

$$\sigma_2' = \sigma_2 h\upsilon, \tag{4}$$

Now the nonlinear absorption coefficient β, is related to the imaginary part of third-order nonlinear optical susceptibility through the equation [6],

$$\text{Im } \chi^{(3)} = n_0^2 \varepsilon_0 c \lambda \beta / 2\pi, \tag{5}$$

Where n_0 is the linear refractive index, ε_0 is the permittivity of free space and c is velocity of light in vacuum.

From the open aperture Z-scan data, the measured values of nonlinear absorption coefficient, β, molecular TPA cross sections σ_2 and σ_2', and the imaginary part of third-order nonlinear optical susceptibility $\chi^{(3)}$ of the liquid crystal sample are given in Table 1. The value σ_2' of liquid crystal samples is nearly two orders of magnitude larger than that of Rhodamine 6G, which is 10^{-48} to 10^{-50} cm^4 s/photon [13]. It is also comparable with the values obtained in stilbazolium like dyes such as trans-4-[2-(pyrrl) vinyl]-1-methylpyridium iodide [14].

Table 1. Third-order nonlinear optical parameters of banana shaped liquid crystals

n_2 ($\times 10^{-11}$ esu)	β (cm/ GW)	Re χ^3 ($\times 10^{-13}$ esu)	Im χ^3 ($\times 10^{-13}$ esu)	σ_2 ($\times 10^{-19}$ cm^4/ GW)	σ_2' ($\times 10^{-46}$ cm^{4}s/ photon)
−1.283	2.232	−1.638	0.446	3.705	1.381

To determine the sign and magnitude of nonlinear refraction, closed-aperture Z-scan was performed by placing an aperture in front of the detector (closed aperture). The closed aperture Z-scan curve is shown in Figure 3(b). To obtain a pure nonlinear refraction curve we adopt the division method described in [6]. The liquid crystalline sample was found to exhibit peak–valley characteristic, indicating negative nonlinear refraction or self- defocusing effect shown in Figure 3(c). The nonlinear refractive index γ (m^2/W) is given by the formula [6],

$$\gamma = \frac{\Delta\phi_0 \lambda}{2\pi L_{\text{eff}} I_0} \, (m^2/W), \tag{6}$$

where $L_{\text{eff}} = \frac{(1-\exp^{-\alpha L})}{\alpha}$ and $\Delta\Phi_0$ is the on-axis phase change given by the equation,

$$\Delta\phi_0 = \frac{\Delta T p - v}{0.406(1 - S)^{0.25}} \quad \text{for } |\Delta\phi_0| \leq \pi, \tag{7}$$

Here ΔT_{p-v} is the peak to valley transmittance difference and S is the linear aperture transmittance. Then nonlinear refractive index n_2 (in esu) is related to γ (m^2/W) by,

$$n_2 \text{ (esu)} = (cn_0/40\pi)\gamma \text{ (m}^2/\text{W)}, \tag{8}$$

The normalized transmittance for pure nonlinear refraction is given by [6],

$$T(Z) = 1 - \frac{4x\Delta\phi_0}{[(x^2 + 9)(x^2 + 1)]}, \tag{9}$$

The nonlinear refractive index n_2 (esu) is related to the real part of third-order nonlinear optical susceptibility through equation [6],

$$\text{Re } \chi^{(3)} = 2n_0^2 \varepsilon_0 c n_2 (esu), \tag{10}$$

Where n_0 is the linear refractive index, ε_0 is the permittivity of free space and c is velocity of light in vacuum.

From the pure nonlinear refraction Z-scan data, the real part of third-order non-linear optical susceptibility $\chi^{(3)}$ and nonlinear refractive index n_2, for liquid crystal sample have been calculated to be given in Table 1.

4. Conclusions

In summary, third-order nonlinear optical parameters of banana shaped liquid crystal in DMF solution were studied using Z-scan technique. This shows a good third-order nonlinear optical response. The nonlinear index found to be negative and magnitude is of the order of 10^{-11} esu. We also measured the molecular two-photon absorption cross-section of this molecule and it is of the order of 10^{-46} cm^4 s/photon which is nearly two orders of magnitude large compared to that of Rhodamine dyes. Hence, the banana shaped liquid crystal investigated seems to be promising candidates for future photonic and optoelectronic applications.

Acknowledgment

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