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Z-can studies and LDT analysis of an organic single crystal: 1-(4-Nitrophenyl) pyrrolidine

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Abstract: A single crystal of 1-(4-Nitrophenyl) pyrrolidine was grown by slow evaporation solution growth technique. The estimations of third order non-linear optical properties like non-linear absorption coefficient (β), non-linear refractive index (n_2) and susceptibility [$\chi^{(3)}$] have calculated using Z-scan technique and these results indicate that the crystal exhibits saturation absorption and self-defocusing performance. The laser damage threshold of the crystal was 2.18 GW/cm^{-2} has been measured by irradiating the (001) crystal plane using a Q-switched Nd: YAG laser.
Keywords: Organic compound, Crystal Growth, X-ray diffraction, nonlinear optics, LDT.

1. Introduction

In recent years, the interest in organic complexes has been widely applied due to a great extent to the possibility of tailoring their nonlinear response by manipulation of their chemical structure and state of aggregation [1]. The search for new materials with high optical nonlinearities has been the important task, because of their practical applications in optical switching, optical bistability and optical limiting, phase modulation and other signal processing [2-3]. To assess a material for the above application, the material must meet several specifications such as high optical nonlinearity, a broadband response, a high threshold for laser induced damage and a low intensity threshold to activate the nonlinear process. Optical-grade organic single crystals of substituted pyrrolidine derivatives with high optical nonlinearities and low melting temperatures are promising materials for optoelectronics, and nonlinear optical (NLO) applications [4]. In this paper, we are reporting the three important parameters of the third order nonlinear optical applications such as nonlinear linear refractive index, nonlinear absorption coefficient, and laser damage threshold [LDT].

2. Synthesis and Growth

4NPY have prepared by following the literature [5]. The calculated amounts of pyrrolidine, Na_2CO_3, 2-propanol has combined with 4-chloronitrobenzene with water. The mixture has refluxed for 1 hour. The resultant mixture has filtered washed with cold water and dried in vacuo at 50°C. The final product has purified by recrystallization process using ethanol as solvent. In order to grow the single crystals of 4NPY, a saturated solution have prepared at room temperature and placed in a constant temperature bath. When solution evaporates, the saturation leads to nucleation and the growth of the crystals. The grown crystal is shown in Fig. 1(a).

3. Results and discussion

3.1. Single crystal XRD

The single crystal X-ray diffraction (XRD) analysis of 4NPY crystal have carried out using ENRAF NONIUS X-ray diffractometer with MoK_α radiation of wavelength λ=0.71073Å at room temperature. The compound crystallized in orthorhombic space group Pbca. The obtained lattice parameters are a = 10.3270 (5) Å, b = 9.9458 (6) Å, c = 18.6934 (12) Å, α =β = γ=90°. Supplementary data have been deposited with the Cambridge Crystallographic Centre, CCDC No. 957066 for the title compound.

3.2 Z-scan studies

The single beam Z-Scan is a well known technique for measuring the degenerate nonlinearities introduced by Sheik-Bahae *et al*., in 1990 [6]. It allows the measurement of both the nonlinear refractive index and nonlinear absorption coefficient. The third order NLO properties have estimated by Z-scan technique with a He-Ne laser (632.8 nm) with the sample thickness 2mm. The Gaussian laser beam was focused by a convex lens having a focal length 30mm and the radius of the aperture is 1mm. The effective thickness (L_{eff}) of 4NPY crystal is 1.93mm. In this method, the sample has translated in the Z-direction along the axis of a focused Gaussian beam and the far field intensity has measured as a function of the sample position. The sample causes an additional focusing and defocusing, depending on whether nonlinear refraction is positive or negative. The sensitivity to nonlinear refraction is entirely due to the aperture, and the removal of aperture completely eliminates the effect. The enhanced transmission near the focus is suggestive of the saturation of the absorption at a high intensity. Absorption saturation in the sample enhances the peak and decreases the valley in the closed aperture Z-scan. The difference between the peak and valley transmission (ΔT_{p-v}) has written in terms of the on-axis phase shift $|\Delta\varphi|$ at the focus. From the closed aperture Z-scan data, the nonlinear refractive index (n_2) have estimated from the relation, $n_2 = \Delta\varphi/kI_0L_{eff}$, Where k is the wave number (k=2π/λ), I_0 is the intensity of the laser beam at the focus (Z=0) and the effective thickness of the sample has calculated using the relation L_{eff} = 1-exp (-αL)/α, Where 'α' is the linear absorption and $L_{eff,}$ is the thickness of the sample. From the open aperture Z-scan data, the nonlinear absorption coefficient (β) has estimated from the relation, $\beta = 2\sqrt{2}\,\Delta T/I_o\,L_{eff}$, Where ΔT is the one-peak value at the open aperture Z-scan curve, I_0 is the irradiance at the focus. The mechanism of the nonlinear response of crystal is due to the thermal effects. Fig. 1(b) and 1(c) shows the closed and open aperture curve of 4NPY.The real and imaginary parts of the third order nonlinear optical susceptibility $\chi^{(3)}$ have defined as

$$\mathrm{Re}\,\chi^3(esu) = \frac{10^{-4}(\varepsilon_0 c^2 n_0^2 n^2)}{\pi}\left(\frac{cm^2}{W}\right) \quad (1)$$

$$\mathrm{Im}\,\chi^3(esu) = \frac{10^{-2}(\varepsilon_0 c^2 n_0^2 n^2)}{4\pi^2}\left(\frac{cm}{W}\right) \quad (2)$$

Where ε_0 is the vacuum permittivity, n_o is the linear refractive index of the sample and c is the velocity of light in vacuum. For the 4NPY crystal, the open and closed aperture curve shows that the nonlinear absorption (β) is Saturable absorption and nonlinear refractive index (n_2) leads to the self-defocusing. Nonlinear refractive index (n_2) of the 4NPY 1.05145 $\times10^{-11}$ cm^2/W and the value of nonlinear absorption coefficient (8.1580 $\times10^{-5}$m/W) estimated from the open Z-scan curve. The real [Re $\chi^{(3)}$] and imaginary [Im $\chi^{(3)}$] of the third order susceptibility was calculated as 5.326×10^{-6} esu and 2.0777×10^{-6} esu. The values suggest that the crystal may find their applications in solid-state laser design [7].

3.3 Laser damage threshold

The laser damage threshold of material is one of the important parameters that decide the applicability of the material for high power laser applications. A Q-switched Nd: YAG laser operating at 1064 nm radiation was used. The laser was operated at the repetition rate of 10 Hz with the pulse width 30 ns. For the LDT measurements 1 mm diameter beam was focused on the sample with a 30 cm focal length lens. The clear visible damage with audible sound occurred at 171.2 mJ respectively. The laser damage threshold of the crystal was 2.18 GW/cm^2.

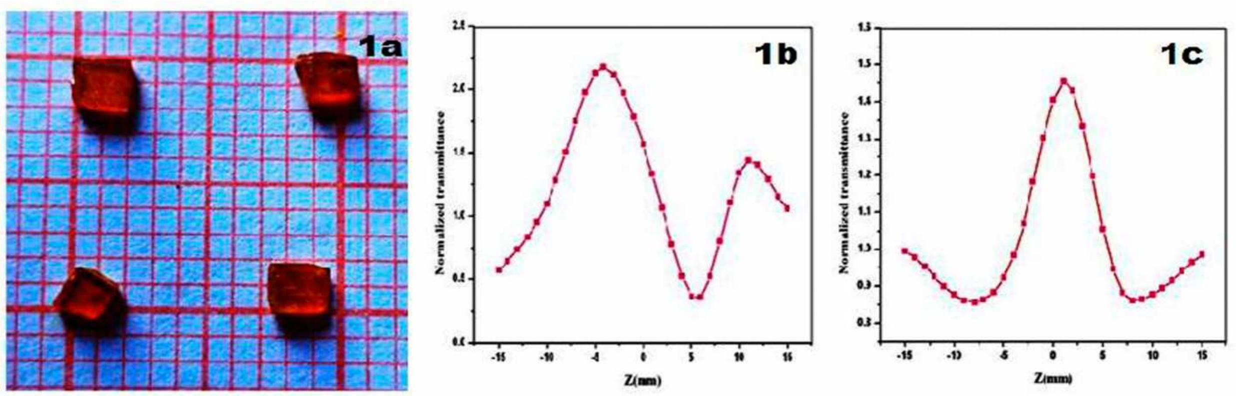

Fig.1 (a) Grown of 4NPY, 1(b) closed aperture curve of 4NPY, (c) open aperture curve of 4NPY

4. Conclusion

Single crystals of 1-(4-Nitrophenyl) pyrrolidine have grown by slow evaporation solution growth technique. The lattice parameters were identified by single crystal x-ray diffraction. Z-scan technique shows that the material exhibits self-defocusing and saturable absorption process. The laser damage threshold of 4NPY crystal is found to be 2.1 GW/cm^2. The study shows that 4NPY crystal is a promising material for third order NLO applications.

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