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Third order Non-Linear optical properties of Ammonium adipate Single Crystals by Z-Scan Technique

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ABSTRACT

We report in this work, Single crystals of Ammonium adipate were grown by the solution growth method using de-ionized water as a solvent. Crystal up to the size of $1.5 \times 0.9 \times 1.9\ cm^3$ *was grown for optical characterization. Optical quality of crystal was observed to be good. The grown crystals were characterized by X-ray diffraction methods. The nonlinear refractive index* n_2 *and susceptibility* $\chi^{(3)}$ *have been measured through the Z-scan technique. Ammonium adipate exhibits saturation absorption and self-focusing performance. Non-linear absorption co-efficient* β *is determined as* $9.5721\times10^{-6}\ cm/W$. *Non-linear refractive index* n_2 *measured at the wavelength of 632.8nm is calculated as* $6.8463 \times 10^{-8}\ cm^2/W$. *The real and imaginary parts of* $\chi^{(3)}$ *have been measured at 632.8 nm and were found to be* $4.6687 \times 10^{-6}\ esu$ *and* $3.4875 \times 10^{-7}\ esu$ *respectively. Also, the absolute value of the third order Non-linear optical susceptibility* $\chi^{(3)}$ *is* $4.6817 \times 10^{-6}\ esu$.

Keywords: Growth from solution, ammonium adipate, single XRD, Z-Scan.

INTRODUCTION

Nonlinear optics and photonics are important disciplines for the development of high-tech applications in many research fields and industries. The study and development of new optical materials have therefore attracted the interest of several research groups. Nonlinear optical processes, resulting from the interaction of laser beams with matter are particularly relevant since they offer not only the possibility of novel applications, but the opportunity to understand physical processes at the fundamental level. In this context, The Z-Scan technique [1-3] is a popular method for the measurement of optical non-linearity of the material. It has the advantage of high sensitivity and simplicity. In the Z-scan technique, the transmittance of a tightly focused Gaussian beam through a finite aperture in the far field is measured as a function of the sample position 'z' with respect to the focal plane. At each position, the sample experiences a different light intensity. The nonlinear refraction of the sample causes a spatial beam broadening or narrowing in the far field and thus modifies the fraction of light that passes through the aperture as the sample position is changed. A typical peak valley (valley-peak) transmittance curve is obtained when the nonlinear refractive index of the medium is negative (positive). In the limit where the sample can be considered as thin, compared to the beam Rayleigh length $(L<z0)$, it is possible from the peak to valley variation of the measured transmittance curve to evaluate the maximum nonlinear phase-shift and hence, knowing the incident laser power, to obtain the nonlinear refractive index n_2. Removing the aperture in the far field, it is possible to perform nonlinear absorption measurements. Such Z-scan traces are expected to be symmetric with respect to the focal point $(z=0)$ where they exhibit a minimum transmittance in the case of nonlinear absorption (multi-photon absorption) and a maximum for the saturation absorption. For media exhibiting both nonlinear refraction and absorption properties, a closed aperture Z-scan measurement is sensitive to both effects. Dividing the closed aperture data by the open aperture, one yields a Z-scan trace typical of a purely refractive nonlinearity. One can simultaneously measure the magnitude and sign of the non-linear refraction and non-linear absorption, which are associated with the real part $\chi_R^{(3)}$ and imaginary part $\chi_I^{(3)}$ of the third order non-linear susceptibilities. The Z-Scan technique has been used to measure the non-linear optical properties of semiconductors [4,5], dielectrics [6,7] organic or carbon-based molecules [8,9] and liquid crystals [10,11]. In this work, we present the growth of Ammonium adipate single crystals by the solution growth method using de-ionized water as a solvent. Single X-ray diffraction, optical absorption spectrum and Z-Scan measurements were carried out. Z-Scan results reveal that it is a potential candidate for the optical switching [12] and optical limiting [13]. The experimental setup used to measure the nonlinear refractive index and absorption of our crystal sample in this work as depicted in the Figure 1.

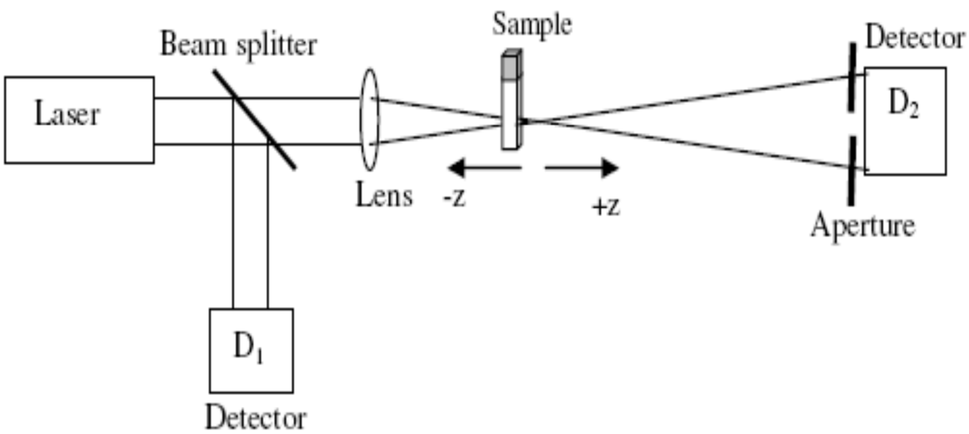

Figure 1 Schematic diagram of Z-scan technique

MATERIALS AND METHODS

CRYSTAL GROWTH

Ammonium adipate crystal was grown using AR grade chemical and de-ionized water as a solvent by slow evaporation technique. The synthesized salt was purified by successive recrystallization process and clear crystals were obtained. The grown ammonium adipate crystal of dimensions $1.5 \times 0.9 \times 1.9\ cm^3$ is shown in Figure 2.

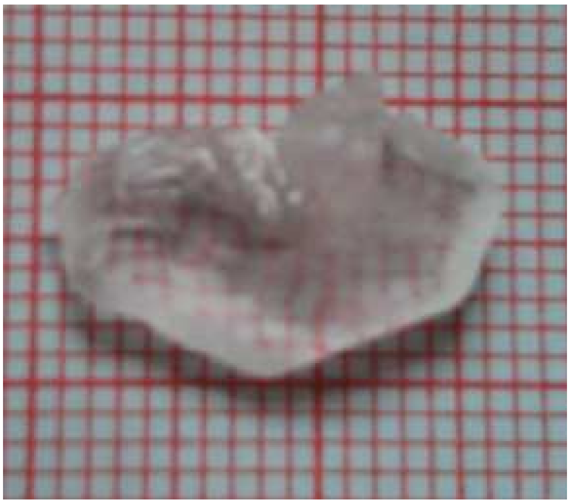

Figure 2 Grown Crystal of Ammonium adipate

RESULTS AND DISCUSSION

3.1 Single Crystal XRD

Single crystal X-ray diffraction analysis for the grown crystals has been carried out using ENRAF NONIUS CAD4 X-Ray diffractometer to confirm the lattice parameters of ammonium adipate. The single crystal data of Ammonium adipate is given in Table 1.

Table 1 The single crystal X-ray data for ammonium adipate

System	Triclinic		
Lattice parameter	$a = 6.152\ \AA$	$b = 8.897(1)\ \AA$	$c = 8.691\ \AA$
	$\alpha = 90°20'$	$\beta = 95°10'$	$\gamma = 100°56'$
Volume (V)	$474.680\ \AA^3$		
Space group	$P2_1/c$		

3.2 NLO MEASUREMENTS

3.2.1. SHG Measurement

Powder SHG studies for ammonium adipate single crystal has been carried out in accordance with the classical powder method developed by Kurtz and Perry [14]. A Q-switched Nd: YAG laser beam of wavelength 1064 nm and pulse width of 8 ns with a repetition rate 10 Hz was used. The ammonium adipate single crystals were powdered with a particle size of around 150 µm and placed in a micro capillary tube and exposed to laser radiation. The second harmonic signal was absent for this sample and it confirms the centro-symmetric nature of the crystals.

3.2.2. Refractive Index Measurement

The refractive index of the ammonium adipate single crystal was determined by Brewster's angle method using He-Ne laser of wavelength 632.8 nm. A polished flattened single crystal is mounted on a rotating mount at an angle varied from 0 to 90 degrees. The angular reading on the rotary stage was observed, when the crystal is perfectly perpendicular to the intra-cavity beam. The crystal was rotated until the laser oscillates and the angle has been set for maximum power output. Brewster's angle (θ_P) for ammonium adipate single crystal is measured to be 57.55^0 (degrees). The refractive index has been calculated using the equation $n = \tan\theta_P$, where θ_P is the polarizing angle and it is found to be 1.595.

3.2.3. Third Order Non-Linear Optical Measurement

A spatial distribution of the temperature in the crystal surface is produced due to the localized absorption of a tightly focused beam propagating through the absorbing sample. Hence a spatial variation of the refractive index is produced which acts as a thermal lens resulting in the phase distortion of the propagating beam. The difference between the peak and valley transmission $\left(\Delta T_{P-V}\right)$ is written in terms of the on axis phase shift at the focus as,

$$\Delta T_{P-V} = 0.406(1-S)^{0.25}\left|\Delta\Phi_0\right| \quad \text{..................... (1)}$$

where, nonlinear phase shift with the sample at focus (Z=0)

$$\Delta\Phi_0 = \frac{2\pi}{\lambda} n_2 I_0 L_{eff} \quad \text{..................... (2)}$$

The nonlinear refractive index is given by

$$n_2 = \frac{\Delta\Phi_0}{K\ I_0\ L_{eff}} \quad \text{..................... (3)}$$

where, $\Delta\Phi_0$ is the phase shift with the sample at focus (Z=0), $K = \frac{2\pi}{\lambda}$ (λ is the laser wavelength), 'L_{eff}' is the effective thickness of the sample $= \frac{\left(1-e^{-\alpha L}\right)}{\alpha}$ 'L' is the thickness of the sample. 'I_0' is the intensity of the laser beam at the focus (Z=0).

"S" is the transmittance of the aperture in the absence of a sample and calculated using the relation

$$S = 1 - \exp\left(\frac{-2\ r_a^2}{\omega_a^2}\right) \quad \text{..................... (4)}$$

where, "r_a" is the aperture and ω_a is the beam radius at the aperture.

From open aperture Z-scan data, the non-linear absorption coefficient is estimated as

$$\beta = \frac{2\sqrt{2}\Delta T}{I_0 L_{eff}} \quad \text{........................ (5)}$$

where, ΔT is the one valley value at the open aperture Z-scan curve.

The value of β will be positive for saturable absorption and negative for two photon absorption. The real and imaginary parts of the third order Non-linear optical susceptibility $\chi^{(3)}$ are defined as

$$\mathrm{Re}\ \chi^{(3)} = \frac{10^{-4}\times\left(\varepsilon_0 c^2 n_0^2 n_2\right)}{\pi} \quad (esu)$$

$$\operatorname{Im} \chi^{(3)} = \frac{10^{-2} \times \left(\varepsilon_0 c^2 n_0^2 \lambda \beta\right)}{4\pi^2} \quad (esu)$$
.... (6)

where, ε_0 is the vacuum permittivity, n_0 is the linear refractive index of the sample and $'c'$ is the velocity of light in vacuum. The absolute value of the third order Non-linear optical susceptibility $\chi^{(3)}$ is calculated from the formula

$$\chi^{(3)} = \sqrt{\left(\operatorname{Im} \chi^{(3)}\right)^2 + \left(\operatorname{Re} \chi^{(3)}\right)^2} \quad (esu)$$
.... (7)

Figure 3 shows the normalized transmittance for the Open Aperture (OA) curve of ammonium adipate. For this case, the transmission is symmetric with respect to the focus (Z = 0), where it has a minimum transmission. This indicates that the sample exhibits Reverse Saturation Absorption (RSA).

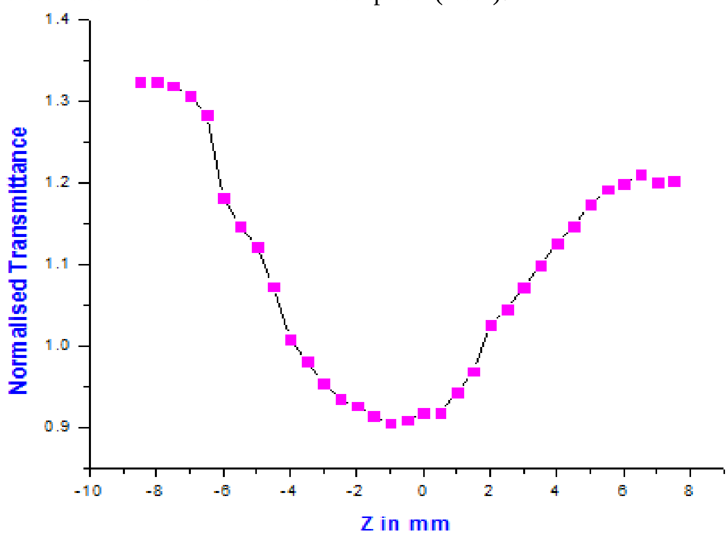

Figure 3 Open Aperture curve of Ammonium adipate

Also, the normalized transmittance for closed aperture curve of (CA) of ammonium adipate is shown in Figure 4. The peak to valley configuration of the curve (figure 4) suggests that the refractive index change is negative, exhibiting a self de-focusing effect. This may be an advantage for the application in protection of optical sensors. As seen from the closed aperture Z –scan curve, the prefocal transmittance peak is followed by the post focal valley which is the signature of negative nonlinearity [15].

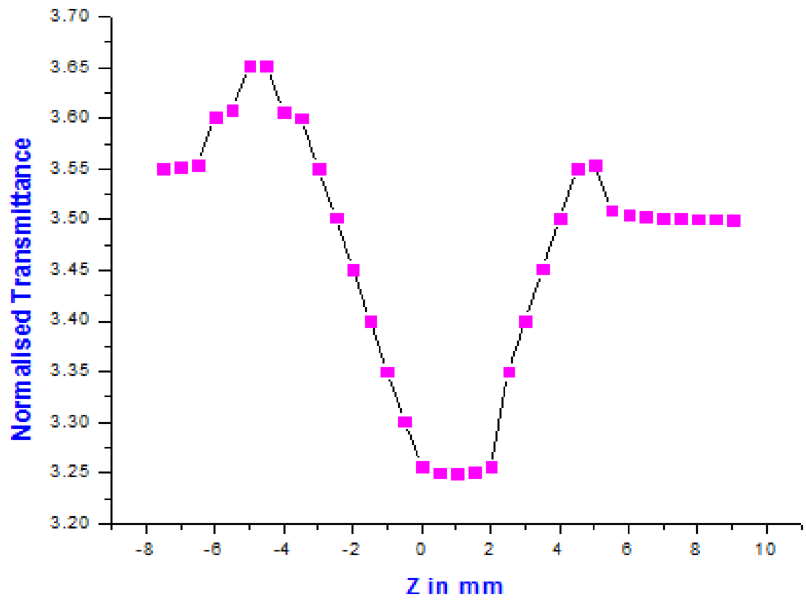

Figure 4 Closed Aperture Curve of Ammonium adipate

Figure 5 shows the division curve of the grown sample by combining OA and CA curves. The division curve is used to calculate third order susceptibility of the grown material.

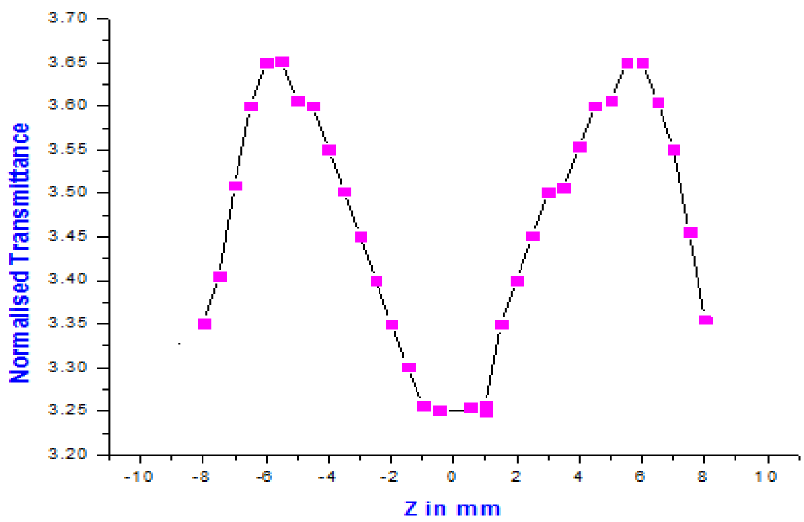

Figure 5 Division Curve of Ammonium adipate

The calculated value of the nonlinear refractive index n_2 is found to be $6.8463 \times 10^{-8} cm^2/W$. As the material has a positive refractive index, it results in self – focusing nature of the material. From the open aperture Z-scan curve, it can be concluded that the nonlinear absorption is regarded as saturation absorption. The nonlinear absorption coefficient (β) is found to be $9.5721 \times 10^{-6} cm/W$. The real and imaginary parts of $\chi^{(3)}$ have been measured at 632.8 nm and were found to be $4.6687 \times 10^{-6} esu$ and $3.4875 \times 10^{-7} esu$ respectively. Also, the absolute value of the third order Non-linear optical susceptibility $\chi^{(3)}$ is $4.6817 \times 10^{-6} esu$. The value of $\chi^{(3)}$ is found to be larger than the other well known compounds [16] and it is due to the p-electron cloud movement from the donor to acceptor which makes the molecule highly polarized. The value of the $\chi^{(3)}$ of ammonium adipate single crystals reported here is of the same order of the magnitude of the materials such as Chaclogenide glasses [17] and C60 [18] etc.

CONCLUSION

We have reported here the optical properties of ammonium adipate single crystal. The absence of SHG efficiency confirms the centro-symmetry nature of the crystal. The Z-scan measurement with 632.8 nm laser pulses revealed that non-linear refractive index of the crystal is in the range of $10^{-8}\,cm^2/W$. The measured third order non-linear properties confirm its suitability for non-linear optical devices such as optical limiting and switching.

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