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Growth and Investigation of the Third Order Non Linear Optical Properties of a Semi Organic Single Crystal Lmhcld

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Abstract— L-Lysine Monohydrochloride Dihydrate (LMHCLDD) single crystals were grown by the slow evaporation method with de-ionized water as a solvent. The grown crystals are of the size 5x 4x1mm^3. The optical properties of the grown crystals are characterized. We need a good optical quality of the crystal is required for the Z-Scan technique. The LMHCLD crystals are having greater figure of merits when comapared to the other crystals in UV-Visual and near IR regions. The unit cell parameters of the grown crystals were studied using single crystal XRD technique. The optical absorption of the grown crystals in the UV– Visual region shows the optical transparency of the grown crystals in the reigon 200 nm to 1400 nm. The second order nonlinear refractive index n_2 and the third order susceptibility $\chi^{(3)}$ of the grown crystals using Z-scan method. From the Z-Scan it is evident that the single crystals of LMHCLD shows the saturation absorption and self-focusing effect. .The higher order nonlinear absorption co-efficient **β** is 8.4113 x 10^{+4} cm^2/W . The calculated non-linear refractive index n_2 for the wavelength 632.8 nm is 1.49 x 10^{-8} cm^2/W. The third order nonlinear susceptibility of the grown crystal is its real part is 1.4438 x 10^{-06} esu and its imaginary parts is around 1.7084 x 10^{-05} esu,

Keywords—(Growth from solution, single XRD, U-V Spectrum, NLO property, Third order Z-Scan)

INTRODUCTION

The Z-Scan technique [1-3]is a highly sensitive, simple and proven technique for measuring the higher order and optical NLO properties of the crystal. The transmittance of a focused laser beam is allowed to pass through a finite aperture. The sample is scanned in the direction of light. The measurements were made at the pre-focal and post focal distances along the direction of light. The light intensity incident on the sample is different at different positions. The light beam passing through an aperture is fractionally modified due to the non linear refraction of the grown crystal with the position of the grown crystal is changed. The grown crystal has positive non linear refractive index. It is observed from its closed aperture curve. For a crystal having positive non linear refractive index, valley to peak transmittance curve is obtained. The condition for the third order nonlinearity is the thickness of the grown crystal is less than the beam Rayleigh Length. i.e. ($L < z_0$) The maximum non-linear phase shift can be evaluated from the peak to valley of the CA transmittance curve. The non linear refractive index n_2 is calculated knowing the incident laser beam power and the peak to valley of CA transmittance curve. The aperture is removed and non-linear absorption measurements were made. These Z-Scan traces are symmetric with respect to the focal point (z = 0), at this point the grown crystals exhibit a minimum transmittance for non linear absorption. (multi photon absorption) It is a maximum for the saturation absorption. The closed aperture reveals that the sample exhibits non linear absorption and nonlinear refraction. The closed aperture data is divided by the open aperture, gives the pure non-linear refractive index of the material chosen. The schematic diagram for Z scan setup is as shown in figure 1.

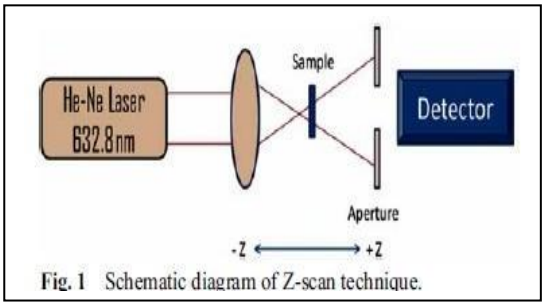

Fig. 1 Schematic diagram of Z-scan technique.

A. CRYSTAL GROWTH

Organic single crystal LMHCLD is grown using the slow evaporation technique using de-ionized water as a solvent The clear crystals were obtained by repeated re-crystallization. The crystals are polished and made to the size of 10 x 5 x 1 mm for Z-Scan studies. The good optical transparency of the grown crystal is as shown in the figure 2.

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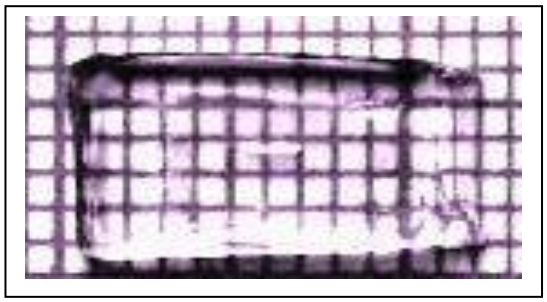

Fig. 2 LMHCLD single crystal

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II RESULTS AND DISCUSSION

Molecular Formula : $C_6H_{14}N_2O_2HCl.2H_2O$

Structural Formula :

Molecular Weight : 182.65 gm/mol

anisotropic, refractive index n = 1.526

Crystal Structure : Monoclinic .

A Single Crystal X ray diffraction

The unit cell parameters of the LMHCLD single crystals are carried out with the ENRAF NONIUS CAD4 X-Ray diffractometer. From the single crystal X- ray Diffraction analysis for the grown crystals the lattice parameters of LMHCLD are obtained. The values are a= 5.887 A°, b= 13.30 A° and c = 7.489 A° and the unit cell volume V=579.80A^3. The grown LMHCLD crystals have the orthorhombic crystal structure. It's crystal symmetry is mm2.[6] LMHCLD posses anisotropic and its growth rate is more along [001] direction than in any other crystal direction. [7].

B. Optical transmission spectrum

The optical transmission spectrum analysis using Perkin Elmer UV-Visual spectrophotometer between 220nm to 800 nm. The recorded spectrum is as shown in the fig 3. The transparency of the grown crystal is observed in the region.

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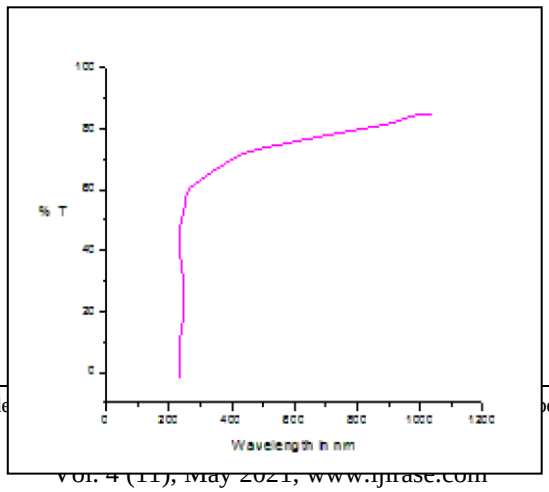


Fig. 3 UV Transmission spectra of LMHCLD..

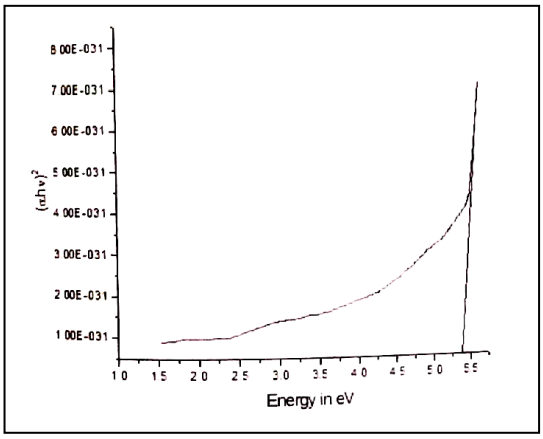
C. Optical bandgap determination

The crystal under study has absorption co-efficient (α) obeying the following relation for the high photon energies. The optical band gap of the grown crystals were calculated using the Tauc's relation

$$\alpha h\nu = (h\nu - E_g)^n$$

where hν is the incident photon energy, ν is the exponent that determines the type of electronic transition causing the absorption and can take the values 1/2 and 2 pending whether transition isdirect or indirect respectively.

Where Eg is the optical band gap of the crystal and A is a constant. The plots variation of $(a\ h\nu)^2$ versus are shown in the figure. Energy gap E_g was evaluated by the extrapolation of the linear part. The band gap energies have been calculated. It's value is 5.4 eV. The tauc plot is as shown in the diagram 4.



D. Refractive index measurement

The refractive index of the grown crystal was determined by the Brewster's amgle method using the gas laser He-Ne of the wavelength 632.8 nm.A polished single crystal is mounted on a rotating mount and rotated through an angle 0 to 90 degree. The crystal mount was rotated till the maximum output is recorded. The angle at which the maximum power opuput recorded is set as the Brewster angle for the grown crystal. . Brewster's angle (θp) is 56.76° for the grown crystal and the refractive index was calculated using the formula n = tan $\boldsymbol{\theta}_p$ polarizing angle for the crystal is found to be 1.526.

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III NLO Measurements

The second order non linear optical properties and third order non linear optical properties of the titled compound were investigated and presented.

A. *SHG Measurements*.

Second Harmonic Generation studies for the grown LMHCLD single crystals were carried out in accordance with the Kurtz Perry method 8]. A Q-switched laser beam of wavelength 1064 nm of pulse width of 8 nano second with a frequency from a Neodymium YAG laser beam was used for the studies. The LMHCLD crystals are powdered to the size of 150 micrometer and filled in a micro capillary tube and exposed to the laser radiations. The second harmonic signal of the sample shows that the SHG efficiency of the LMHCLD is greater than Urea and KDP and L- Arginine etc.

B THIRD ORDER NONLINEAR OPTICAL MEASUREMENT

If a tightly focused beam with the help of a convex lens of 20 cm, localized absorption and a spatial distribution of the incident beam by a grown crystal. It result in the thermal lense effect due to spatial variation of the refractive index and the phase distortion of the propagated beam as a result The valley to peak transmission (ΔTp-v) is found difference in terms of the axis phase shift at the focus

$$\Delta T_{p\text{-}v} = 0.406(1\text{-}S)^{0.25} |\Delta\Phi| \quad \ldots\ldots\ldots\ldots\ldots\ldots(1)$$

The aperture linear transmittance "S" is calculated using the relation

$$S = 1 - \exp(-2r_a^2/\omega_a^2) \quad \ldots\ldots\ldots\ldots\ldots\ldots(2)$$

r_a – radius aperture and w_A the beam radius at the aperture.

The nonlinear refractive index of the LMHCLD crystal is

$$n_2 = \Delta\Phi / K I_0 L_{eff} \quad \ldots\ldots\ldots\ldots\ldots\ldots(3)$$

Where wave vector $K = 2\pi/\lambda$, λ - the laser beam wavelength, The intensity of the laser beam at the forcus is (Z=0),

The effective length L_{eff} =[1-exp(-αd)] of the grown crystal in which we can observe the third order non linear optical properties. "d" the sample thickness. The open aperture Z-scan data, the nonlinear absorption coefficient calculated using the formula

$$\beta = (2\sqrt{2}\,\Delta T / I_0 L_{ef}) \quad \ldots\ldots\ldots\ldots\ldots\ldots(4)$$

The valley transmittance at the open aperture Z-scan curve is ΔT The absorption coefficient β is negative saturation absorption. It confirms the two photon absorption. The third order non-linear optical susceptibility $\chi^{(3)}$; its real and imaginary value is calculated from the formula

$$\chi_{Re}^{(3)} = 10^{-4} (\varepsilon_0 C^2 n_0^2 n_2) / \pi \text{ esu} \quad \ldots\ldots\ldots(5)$$

$$\chi_{Im}^{(3)} = 10^{-2} (\varepsilon_0 C^2 n_0^2 \lambda\beta) / 4\pi^2 \text{ esu} \ldots..(6)$$

The vacuum permittivity is "ε_0"; the linear refractive index of the crystal "n_0" and "c" velocity of light. The open aperture curve of the grown crystal is as shown in the figure 5.

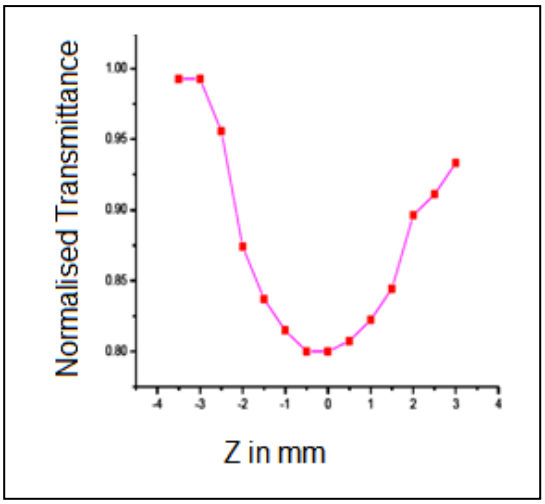

Fig 5 Open Aperture curve of LMHCLD

The optical transmittance is symmetric at the focus (Z=0), where the transmission is minimum confirms the saturation absorption (SA). The closed aperture curve is drawn between the normalized transmittance and the sample position as shown in figure 6. The sample exhibits saturation Absorption (SA).

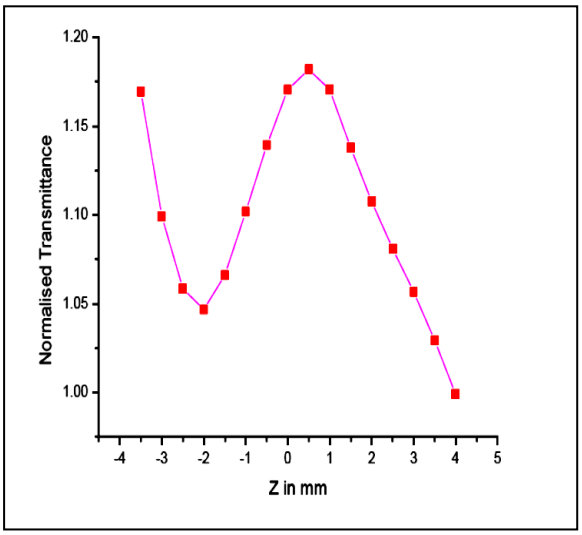

Fig. 5 Closed Aperture Curve of the LMHCLD crystal

From the closed aperture curve (valley to peak configuration of the curve) ; it is evident that the refractive index change is negative. The crystal with negative refractive index confirms the self defocusing effect of the crystal. The grown crystal has the advantage for the application as an optical sensor. Table 2 portrays the experimentally calculated values of the Z –Scan method for LMHCLD crystal From the repeated measurements made on the sample a maximum error bar of about 5% was estimated. The positive nonlinearity (9) obtained from the prefocal transmittance followed by the post focal peak of the closed aperture value of Z – Scan curve. The nonlinear refractive index value calculated as n_2 is 1.49 x 10^{-8} cm^2 /W

The material exhibits self focusing effect due to its positive refractive index nature of the grown crystal. The saturation absorption nature of the material is confirmed from the open aperture Z scan curve of the material. The calculated nonlinear absorption of the grown crystal is ***8.4113 x 10^{-4} cm/W.*** From the Z Scan calculated values of the nonlinear susceptibility of the LMHCLD single crystal is ***2.8088 x 10^{-5} esu***. It is larger than the other well known compounds. [10]. The third order nonlinear susceptibility is due to the movement of π electron cloud movement from the donor to acceptor compounds makes the molecule highly polarized. The third order nonlinear susceptibility value of the LMHCLD crystal is greater than that of the reported materials such as Chaclogenide glasses [11] and C60 [12].

III CONCLUSION

The higher order nonlinear optical properties of the LMHCLD crystal were investigated by Z-Scan technique using the He-Ne laser source of wavelength 632.8 nm were reported. The second harmonic generation efficiency confirms that the LMHCLD is a potential candidate for non-linear optical applications. It's SHG efficiency is greater than the that of Urea, KDP and L-Argentine etc. The investigations reveal that the non-linear refractive indes of the grown crystal is in the range of 10^{-8} cm^2/W. The third order NLO properties confirm the suitability of the grown crystal in the application such as optical switching device, optical gates etc bi-stable optical switching devices, optical power limiters and all optical switching devices etc.

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