

Third order non-linear optical properties of L-asparagine admixed Lithium Chloride (LALC) and Cesium Chloride (LACC) Single Crystals by Z-Scan Technique

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Abstract: Single crystals of LALC and LACC grown by the solution growth method were found to be orthorhombic in structure. Optical quality of crystal was observed to be good. In this present work the nonlinear refractive index (n) and susceptibility (χ) have been measured through the Z-scan technique. This method has been widely used in material characterization because it provides not only the magnitudes of the real part and imaginary part of the nonlinear susceptibility, but also the sign of the real part. Nonlinear absorption co-efficient β is determined, Non-linear refractive index (n) measured at the wavelength of 632.8nm is calculated to be $-2.244 \times 10^{-10} \text{ cm}^2/\text{w}$. From the studies it is inferred that the crystal possess promising third-order optical susceptibilities, which allow to use the crystal as optical limiters.

Keywords: LACC, LALC, Z-Scan.

1. Third-order NLO studies

The Z-scan technique is a popular method for measuring optical nonlinearities using a single laser beam. It is a simple and cheap method to evaluate third-order NLO parameters such as third-order absorption coefficient, third-order refractive index and third-order susceptibility of crystalline samples. A He-Ne laser ($\lambda = 632.8 \text{ nm}$) was used as the light source and focused by a lens of 22.5 cm focal length. The transmitted intensity from the sample was measured as a function of sample position in the Z-direction with respect to the focal plane either through a closed aperture (CA) or open aperture (OA) mode. The open aperture and closed aperture Z-scan curves for LALC and LACC crystalline samples are presented in figures 1 and 2. Using the closed aperture curves, the nonlinear refractive index of the samples was found. Since

the closed aperture curves have peak and then valley format, the LALC and LACC samples have negative third-order refractive index values[1-3]. Open aperture curves for both the samples show normalised transmittance peaks which is due to the result of the presence of negative non linear absorption process. These results indicate that the non linearity is mainly due to the saturable absorption process[4]. Using the open aperture curves, the nonlinear absorption coefficient for the samples was determined. Using both the third-order refractive index and absorption coefficient values, the real part and imaginary part of third-order susceptibility values for LALC and LACC samples were determined and the values are tabulated in table.

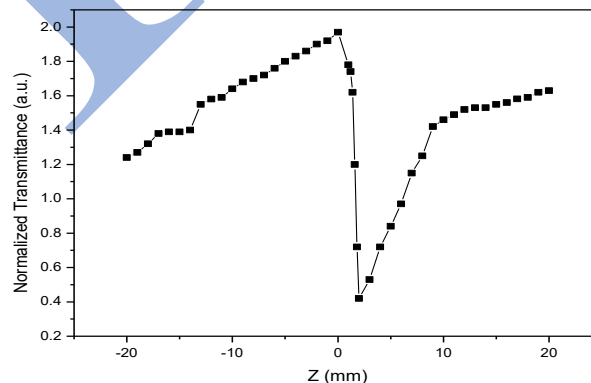


Fig.1.a: Closed aperture Z-scan curve for LALC sample

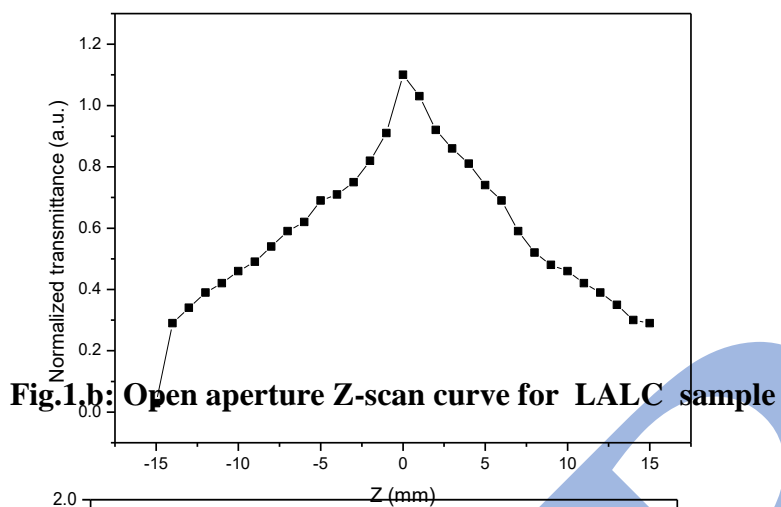


Fig.1.b: Open aperture Z-scan curve for LALC sample

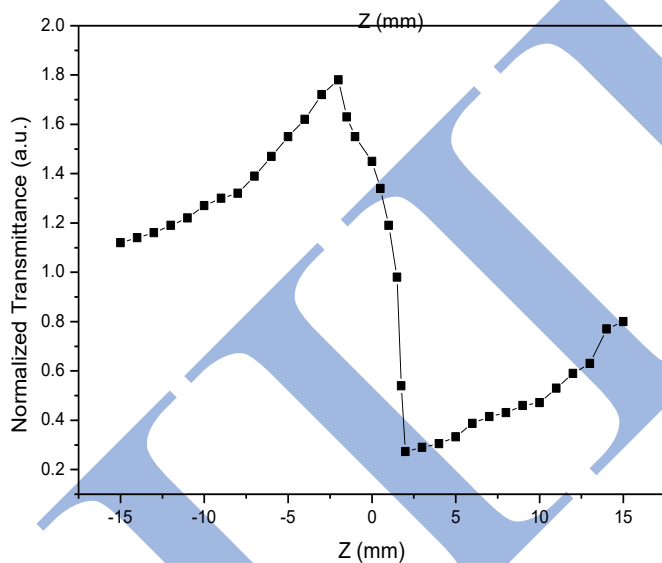


Fig.2.a: Closed aperture Z-scan curve for LACC sample.

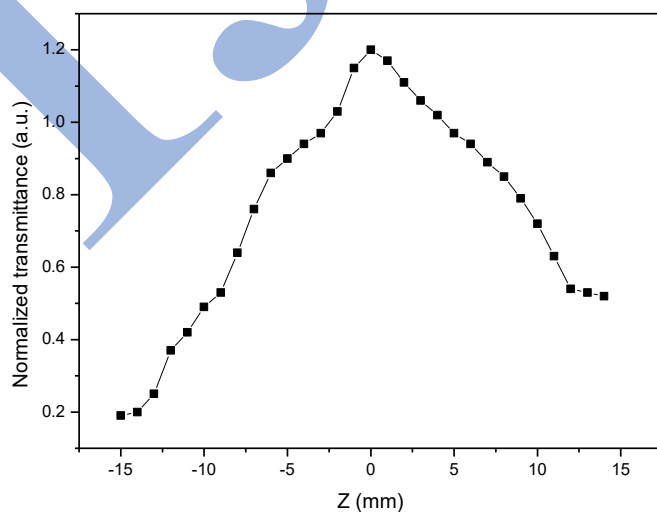


Fig.2.b: Open aperture Z-scan curve for LACC sample.

The transmission difference between peak and valley (ΔT_{p-v}) is written in terms of phase shift

$$\Delta T_{p-v} = 0.406 (1-s)^{0.25} |\Delta\phi|$$

Linear transmittance aperture (S) is calculated using the relation

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

where r_a is the radius of the aperture and ω_a is the beam radius at the aperture. The third order nonlinear refractive index (n_2) of the crystal was calculated by following the relation.

$$n_2 = \frac{\Delta\phi}{KI_0L_{eff}}$$

The effective thickness can be calculated using the relation

$$L_{eff} = [1 - \exp(-\alpha L)] / \alpha$$

The nonlinear absorption coefficient (β) can be calculated using the following relation

$$\beta = \frac{2\sqrt{2}\Delta T}{I_0L_{eff}}$$

$$\text{Real part of } \chi^{(3)} = 10^{-4} (\epsilon_0 c^2 n_0^2 n_2) /$$

$$\pi \quad (\text{esu})$$

$$\text{Imaginary part of } \chi^{(3)} = (10^{-2} \epsilon_0 c^2 n_0^2 \lambda \beta) /$$

$$4\pi^2 \quad (\text{esu})$$

$$|\chi^{(3)}| = \left[(\text{Re } \chi^{(3)})^2 + (\text{Im } \chi^{(3)})^2 \right]^{\frac{1}{2}}$$

When valley comes after a peak in the closed aperture Z-scan diagram, it indicates the signature for negative nonlinearity. This is known as self-defocusing effect which is due to local variation of refractive index with temperature. For materials with multi-photon absorption, there is a minimum transmittance in focus (valley) and for saturable absorber samples, there is maximum transmittance in the focus (peak).

Table : Third-order NLO parameters for LALC and LACC crystals

Third-order NLO parameters	Samples	
	LALC	LACC
Nonlinear refractive index (n_2)	$-2.26 \times 10^{-10} \text{ cm}^2/\text{W}$	$-2.244 \times 10^{-10} \text{ cm}^2/\text{W}$
Nonlinear absorption coefficient (β)	$1.69 \times 10^{-5} \text{ cm/W}$	$1.88 \times 10^{-5} \text{ cm/W}$
Real part of the third-order nonlinear susceptibility	$1.90 \times 10^{-8} \text{ esu}$	$1.885 \times 10^{-8} \text{ esu}$
Imaginary part of the third-order nonlinear susceptibility	$7.179 \times 10^{-7} \text{ esu}$	$8.00 \times 10^{-7} \text{ esu}$
The third-order nonlinear susceptibility ($\chi^{(3)}$)	$0.718 \times 10^{-6} \text{ esu}$	$0.80 \times 10^{-6} \text{ esu}$

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