ABSTRACT

Crystal growth is basically a process of arranging atoms, ions, molecules or molecular assemblies into regular three dimensional periodic arrays. The necessary theories for nonlinear optical phenomena such as second order and third order susceptibilities are explained in lucid manner in the first chapter. The design of nonlinear optical (NLO) molecules has become a focus of current research in view of their potential applications in various photonic technologies. Materials with NLO activity find use as electro-optic switching elements for telecommunication and optical information processing.

The second chapter deals the principle and working mechanism of various characterization techniques involved in the present investigation in detail.

The third chapter deals the electro-optical modulator and third order nonlinearity of an organic single crystal of Imidazolium L-tartrate (IMLT) <010> grown by unidirectional growth method of Sankaranarayanan-Ramasamy (SR). The grown IMLT seed crystals were characterized by single crystal XRD and the lattice parameters were confirmed. The UV-Visible transmittance analysis shows that SR method grown IMLT single crystal possesses good transmittance in the entire visible region with a low cut-off wavelength at 240 nm. The mechanical study on the (010) plane of the IMLT single crystal analyzed by Vickers microhardness tester reveals the moderate hardness of the material. Refractive indices of the orthogonally cut and polished IMLT single crystal were determined by Brewster angle method. Second Harmonic Generation (SHG) efficiency of IMLT is 4.3 times greater than the standard Potassium Dihydrogen Phosphate (KDP) crystal. Nonlinear

refractive index (n_2 = 1.853 × 10^{-8} cm^2/W), Nonlinear absorption coefficient (β = 6.1801×10^{-4} cm/W) and Third-order nonlinear optical susceptibility $\chi^{(3)}$ = 6.6579 × 10^{-6} esu were evaluated using Z-scan method. The half wave voltage (3.1 kV) and electro-optical coefficient (66.19 pm/V at 532 nm) of IMLT crystal were determined using polarimetric technique. The electro-optical coefficient (r_{12}) of IMLT is found to be greater than that of KDP crystal (r_{41} = 8.77 pm/V) which represents the suitability of IMLT crystal to device the electro-optical modulator.

The fourth chapter deals with the evaluation of suitability of ammonium D, L-Tartrate (AMT) single crystal for optical limiting application by performing structural, dielectric, mechanical, optical and third order nonlinear characterization studies. The AMT was grown by conventional solution growth technique. The unit cell parameters were obtained from single crystal XRD analysis and the crystal system is confirmed to be orthorhombic with noncentrosymmetric space group $P2_12_12_1$. The crystalline perfection was evaluated by high resolution X-ray diffractometry (HRXRD). The optical transparency window from 234 nm to 1100 nm region was analyzed by UV – Visible spectral studies. Dielectric measurements were made over a wide range of frequencies (5 kHz to 2 MHz) and temperature to analyze real dielectric constant and dielectric loss of the AMT single crystal. The SHG efficiency of AMT is found to be 1.3 times greater than the standard KDP crystal. From the open aperture Z - scan curve the value of nonlinear absorption coefficient β is found to be 3.6685 × 10^{-4} cm/W. The closed aperture Z-scan curve reveals the negative nonlinear refractive index and the value is 2.1096 × 10^{-8} cm^2/W and the third order susceptibility $\chi^{(3)}$ = 6.71 × 10^{-6} esu.

Comparative studies on unidirectional method grown organic Benzimidazole (BMZ) and semiorganic Potassium Sodium Tartrate (PST) single crystals for efficient optical limiting applications are dealt in the fifth

chapter. Single crystal X - ray diffraction analysis reveals that BMZ belongs to orthorhombic crystal system with space group $Pna2_1$ and PST belongs to orthorhombic crystal system with centrosymmetric space group $P2_12_12$.

The crystalline perfection of cut and polished wafers of 2 mm thick BMZ and PST were examined by high resolution X - ray diffractometry. From this analysis it is found that the quality of the grown crystal is quite good. UV-Visible analysis shows the optical absorption cut – off wavelength of 1 mm thick BMZ and PST single crystals. Dielectric measurements were made over a wide range of frequencies and temperature to analyze real dielectric constant and dielectric loss. The second harmonic generation efficiency of both the materials were studied by Kurtz – Perry test and compared with potassium dihydrogen phosphate (KDP). Open aperture Z-scan experiment was performed to observe absorptive nonlinearity. The value of nonlinear absorption coefficient (β) is found to be 159.99×10^{-12} m/W, 100×10^{-12} m/W and 37.5×10^{-12} m/W, respectively for BMZ, PST and KDP crystals. Optical limiting properties were studied by keeping the sample at the focus and the results were compared.

The sixth chapter discusses the growth, mechanical, dielectric, linear and nonlinear optical behaviour of slow evaporation solution grown Zinc guanidinium sulphate (ZnGuS) single crystal. The single crystal XRD study confirms the ZnGuS belongs to the tetragonal crystal system. The UV – Visible transmittance and absorption spectra show the low cut – off wavelength (230 nm) and wide transparency. Hardness study shows the reverse indentation size effect. The particle size dependent SHG efficiency of ZnGuS was analyzed and compared with standard KDP. Nonlinear absorption $\beta = 7.5834 \times 10^{-4}$ cm/W and nonlinear refractive index $n_2 = 6.377 \times 10^{-9}$ cm^2/W and third order susceptibility $\chi^{(3)} = 0.3018 \times 10^{-6}$ esu were calculated by employing experiment using Nd:YAG laser SHG output wavelength of 532 nm. Optical limiting property was studied by placing the sample at focus.

The seventh chapter covers the optical, dielectric and third order nonlinear optical properties of <011> orientation unidirectional method grown L – Histidine Nitrate (LHN) single crystal. The single crystal XRD study confirms the LHN crystal belongs to the orthorhombic crystal system with noncentrosymmetric space group $P2_12_12_1$. The cell parameter values are a = 5.239 (5) Å, b = 7.081 (1) Å, c = 24.96 (1) Å and V = 926.01 $Å^3$. $\alpha = \beta = \gamma = 90^{\circ}$. The lower cut – off wavelength was observed at 320 nm in the UV – Visible transmittance spectrum. The dielectric constant and dielectric loss were studied with different frequency and various temperatures. The SHG efficiency of LHN with different particle size was analyzed. Nonlinear refractive index ($n_2 = 1.2057 \times 10^{-8}$ cm^2/W), nonlinear absorption coefficient ($\beta = 3.482 \times 10^{-4}$ cm/W) and third-order nonlinear optical susceptibility $\chi^{(3)} = 0.238 \times 10^{-6}$ esu were evaluated using Z- scan method. The optical limiting property of LHN was assessed.

The eighth chapter discusses the comparative studies on optical, dielectric, laser damage threshold, third order susceptibility and optical limiting behaviour of Czochralski method grown 1 mole %, 4 mole % and 5 mole % MgO doped Lithium Niobate (LN) crystals. UV – Visible transmittance and absorption spectra were recorded. Third order nonlinear susceptibility and optical limiting behaviour were analysed and compared. The value of nonlinear absorption coefficient β of 1 mole %, 4 mole % and 5 mole % MgO doped Lithium Niobate is 1.8017×10^{-4} cm/W, 3.181×10^{-5} cm/W and 7.5834×10^{-4} cm/W, respectively. The nonlinear refractive index is 6.377×10^{-9} cm^2/W, 1.910×10^{-9} cm^2/W and 1.194×10^{-9} cm^2/W, respectively for 1 mole %, 4 mol % and 5 mol % MgO doped $LiNbO_3$.

The conclusions drawn out of the present investigation and the suggestions for future work are presented in ninth chapter.