

## ACKNOWLEDGEMENT

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(SATYA KUMAR KUSHWAHA)

## PREFACE

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The modern era of science is facing a challenge of managing the huge data of Astronomers with huge telescopic images, high-energy physicist, and bioinformaticians with vast biological databases for its storage, execution and transmission. The perfect single crystals of photorefractive *nonlinear optical* (NLO) materials are most suitable to overcome this information overload to a great extent and in the past decades the efforts are made to store and read-out the information with higher capacities and rates, in the form of *holograms*. Storing the information holographically is just one aspect of a move towards making faster, more powerful computer systems that *think* using light rather than electricity with the use of mighty laser beams. The NLO materials with high optical efficiencies have opened a window for the development of lasers with tunable energies required for inertial confinement fusion research and optical display and have been the centre of attraction for research and development. The series of new inorganic, organic and semi-organic NLO materials have been discovered and their single crystals have been developed. Lithium Niobate ( $\text{LiNbO}_3$ ) termed as *silicon of photonics* is indispensable in advanced photonics and nonlinear optics and has been extensively studied.

The research work carried out in the thesis concerns growth and investigation of crystalline perfection *vis-à-vis* physical properties of device quality NLO single crystals and the following problems have been investigated:

- (i) Czochralski growth, crystalline perfection and optical characterization of pure and Zn-doped  $\text{LiNbO}_3$  NLO single crystals.
- (ii) Crystalline perfection and optical properties of Czochralski grown Fe-doped  $\text{LiNbO}_3$  in view of photorefractive NLO applications.
- (iii) Enhancement in crystalline perfection and optical properties of Benzophenone single crystals: A remarkable effect of liquid crystal.
- (iv) A correlation of crystalline perfection with enhancement of SHG efficiency by urea doping in ZTS single crystals.
- (v) Effect of  $\text{Cr}^{3+}$ -doping on crystalline perfection and optical properties of ZTS.

The present thesis is divided into eight chapters. *Chapter 1* gives a brief survey of the NLO single crystals and their technological importance. The theory of

nonlinear optics; second harmonic generation, phase matching, has been reviewed. The theory of nucleation, subsequent crystal growth theory and crystal growth techniques have been reviewed. The commonly observed structural defects in the single crystals have also been discussed. *Chapter 2* describes the theory and experimental details of Czochralski (CZ) and solvent evaporation growth methods along with the details about the *indigenously* developed *Czochralski pullers* and the processes used for growth of pure and doped NLO single crystals of LiNbO<sub>3</sub>, Benzophenone and ZTS (tris(thiourea)zinc sulphate) have been provided. In *Chapter 3* all the characterization techniques along with their fundamentals/principles, used for the characterization of grown crystals have been discussed in detail. Special emphasis has been given on the *in-house* developed *multicrystal high resolution X-ray diffractometer* (HRXRD) used for crystalline perfection investigations.

*Chapter 4* illustrates the CZ growth of pure and Zn-doped LiNbO<sub>3</sub> bulk single crystals and their characterization by powder X-ray diffraction, HRXRD, Raman, UV-VIS-NIR, Fourier transform infrared (FTIR), prism coupler (PC) and ellipsometry for the investigation of crystalline perfection and various optical parameters. *Chapter 5* demonstrates the growth of a photorefractive Fe-doped LiNbO<sub>3</sub> NLO single crystal by CZ method. Characterization of crystal for crystalline perfection, ionic state of Fe, optical transparency/band gap, Raman scattering modes, proton incorporation into lattice, birefringence and wavelength dispersion of linear refractive index has been carried out. *Chapter 6* discusses the influence of *liquid crystals* (LC) (added to the charge during growth) on crystalline perfection and optical transparency of Benzophenone (BP). *Chapter 7* describes the enhancement in second harmonic generation (SHG) efficiency of ZTS with urea doping and its correlation with crystalline perfection. *Chapter 8* describes the influence of Cr<sup>3+</sup> doping on crystalline perfection, optical transparency/band gap and linear refractive index of ZTS single crystals by employing HRXRD, photoluminescence, UV-VIS-NIR and ellipsometry techniques.