

CONTENTS

ACKNOWLEDGEMENT	i
PREFACE	iii
1 Literature survey and theoretical aspects of nonlinear optics, crystal growth and lattice imperfections	1
Abstract	1
1.1 LITERATURE SURVEY	2
1.2 THEORETICAL ASPECTS OF NONLINEAR OPTICS	7
1.2.1 Nonlinear optics	7
1.2.2 Harmonic generation	7
1.2.3 Second harmonic generation	9
1.2.4 Phase matching	13
1.2.5 Phase matching techniques	13
1.2.5.1 Birefringence phase matching	14
1.2.5.2 Beam <i>walk-off</i> and temperature tuning for 90° phase-matching	16
1.2.5.3 Quasi phase matching by periodic poling	18
1.3 THEORETICAL ASPECTS OF CRYSTAL GROWTH	21
1.3.1 Theory of crystallization	22
1.3.2 Nucleation	24
1.3.3 Subsequent growth of nuclei	26
1.3.3.1 Two dimensional layer growth theory	26
1.3.3.2 Screw dislocation theory	27
1.4 SINGLE CRYSTAL GROWTH METHODS	27
1.4.1 Melt growth	28
1.4.2 Solution growth	29
1.4.3 Vapour phase growth	30
1.5 IMPERFECTIONS/ DEFECTS IN SINGLE CRYSTALS	30
1.5.1 Grain boundaries	31
1.5.2 Low angle or subgrain boundaries	31
1.5.3 Dislocations	32

1.5.4	Stacking faults	32
1.5.4	Point defects and their aggregates	33
2	Experimentation for the growth of single crystals	35
	Abstract	35
2.1	CZOCHRALSKI CRYSTAL GROWTH	36
2.1.1	Salient features of Czochralski technique.	36
2.1.2	Czochralski growth of pure, Zn- and Fe-doped LiNbO ₃ single crystals	39
2.1.2.1	Phase diagram of LiNbO ₃	39
2.1.2.2	Bulk crystal growth	41
2.1.3	Czochralski growth of pure and liquid crystal induced Benzophenone single crystals	48
2.1.2.1	Materials properties and processing	48
2.1.2.2	Bulk crystal growth	48
2.2	SOLUTION CRYSTAL GROWTH	50
2.2.1	Salient features of solution growth	50
2.2.2	Meta-stable zone width and solubility curve	51
2.2.3	Induction period and nucleation kinetics	52
2.2.4	SEST growth of pure, Urea and Cr ³⁺ doped ZTS single crystals .	53
2.2.4.1	Materials synthesis process	53
2.2.4.2	Single crystal growth	54
2.3	CONCLUSION.	55
3	Characterization techniques	57
	Abstract	57
3.1	ATOMIC ABSORPTION SPECTROSCOPY (AAS)	58
3.2	POWDER X-RAY DIFFRACTION (PXRD)	59
3.3	HIGH RESOLUTION X-RAY DIFFRACTOMETRY (HRXRD)	62
3.4	FT-IR SPECTROSCOPY	67
3.5	RAMAN SPECTROSCOPY	69
3.6	NUCLEAR MAGNETIC RESONANCE SPECTROSCOPY	71
3.7	EPR SPECTROSCOPY	73

3.8	PRISM COUPLER SPECTROMETER	74
3.9	ELLIPSOMETRY	76
3.10	POLARIZED OPTICAL MICROSCOPY	78
3.11	PHOTOLUMINESCENCE SPECTROSCOPY	80
3.12	UV-VIS-NIR SPECTROSCOPY	81
3.13	KURTZ POWDER SHG TECHNIQUE	83
4	Czochralski growth, crystalline perfection and optical characterization of LiNbO₃ and Zn:LiNbO₃ NLO single crystals	87
	Abstract	87
4.1	INTRODUCTION	88
4.2	CRYSTAL GROWTH	90
4.3	CHARACTERIZATION STUDIES	93
4.4	RESULTS AND DISCUSSION	94
4.4.1	Powder X-ray diffraction analysis	94
4.4.2	High resolution X-ray diffraction analysis	96
4.4.3	Raman spectroscopy analysis	100
4.4.4	Fourier transform infrared analysis	102
4.4.5	UV-VIS-NIR optical analysis	105
4.4.6	Prism coupler birefringence analysis	109
4.4.7	Wavelength dispersion analysis	111
4.5	CONCLUSION	114
5	Crystalline perfection and optical properties of Czochralski- grown Fe:LiNbO₃: A photorefractive NLO single crystal	117
	Abstract	117
5.1	INTRODUCTION	118
5.2	CRYSTAL GROWTH	120
5.3	CHARACTERIZATION STUDIES	122
5.4	RESULTS AND DISCUSSION	123
5.4.1	Powder X-ray diffraction analysis	123

5.4.2	High resolution X-ray diffraction analysis	125
5.4.3	EPR analysis	127
5.4.4	Raman spectroscopic analysis	128
5.4.5	Fourier transform infrared analysis	130
5.4.6	UV-VIS-NIR analysis	130
5.4.7	Prism coupler birefringence analysis	133
5.4.8	Wavelength dispersion analysis	135
5.5	CONCLUSION	137

6 Enhancement in crystalline perfection and optical properties of

Benzophenone single crystals: A remarkable effect of

liquid crystals **139**

Abstract 139

6.1	INTRODUCTION	140
6.2	CRYSTAL GROWTH	142
6.3	CHARACTERIZATION STUDIES	142
6.4	RESULTS AND DISCUSSION	144
6.4.1	Powder X-ray diffraction analysis	144
6.4.2	Nuclear magnetic resonance analysis.	145
6.4.3	High-resolution X-ray diffraction analysis	146
6.4.4	Photoluminescence emission analysis	150
6.4.5	UV-VIS-NIR optical analysis	151
6.4.6	Polarized optical microscope birefringence analysis	152
6.5	CONCLUSION	154

7	A correlation of crystalline perfection with SHG efficiency of urea doped ZTS single crystals	155
	Abstract	155
7.1	INTRODUCTION	156
7.2	CRYSTAL GROWTH	159
7.3	CHARACTERIZATION STUDIES	160
7.4	RESULTS AND DISCUSSION	161
7.4.1	Fourier transform infrared analysis	161
7.4.2	Powder X-ray diffraction analysis	163
7.4.3	High resolution X-ray diffraction analysis	164
7.4.3.1	Undoped specimen	166
7.4.3.2	Specimens doped with concentrations up to 2.5 mol%	168
7.4.3.3	Specimens doped with concentrations between 5 to 12 mol%	172
7.4.4	SHG efficiency analysis	176
7.5	CONCLUSION	177
8	Effect of Cr³⁺-doping on crystalline perfection and optical properties of ZTS: The NLO single crystals	179
	Abstract	179
8.1	INTRODUCTION	180
8.2	CRYSTAL GROWTH	181
8.3	CHARACTERIZATION STUDIES	181
8.4	RESULTS AND DISCUSSION	182
8.4.1	Atomic absorption spectroscopic and - powder X-ray diffraction analysis	182
8.4.2	High resolution X-ray diffraction analysis	184

8.4.3	Photoluminescence analysis	187
8.4.4	UV-VIS-NIR analysis	188
8.4.5	Wavelength dispersion analysis	190
8.5	CONCLUSION	194
	REFERENCES	195
	SUMMARY AND FUTURE SCOPE OF WORK	213
I.	SUMMARY	213
II.	FUTURE SCOPE OF WORK	215
	LIST OF PUBLICATIONS, CONFERENCE AND AWARDS	217