Crystalline solids exhibit highest order of molecular arrangement among the materials. Ideal crystals contain infinite lattice of regularly arranged atoms in three dimensions. Most of the crystals that we encounter are finite in structure and may contain defects. However, the unique properties they possess make them different from other solids. With the advancement of technology, crystalline forms of matter found applications in different areas of human activity. This, in turn enhanced the demand for crystals with novel properties and the researchers have developed many techniques for producing them artificially.

The perfection, external form and homogeneity of an artificial crystal are related, by and large, to the method employed for its growth. Nowadays, artificially grown crystals find unbelievable and innumerable applications in scientific research, semiconductor industry, microelectronics, medical instrumentation, communication technology, military regime, energy and space technology, bearing systems and jewellery. Quite obviously, the responsibility of developing novel crystals with hitherto unknown properties falls on the shoulders of solid state physicists.

There are four vital stages in the growth of a crystal from an initially disordered phase; attainment of a supersaturated phase, formation of crystal nuclei of microscopic size, subsequent growth of nuclei into crystals with distinct faces and cessation of growth. Nucleation is considered as the precursor of the crystallization processes. Once the critical size is attained, further addition of molecules takes place in a regular manner which is characteristic of the lattice and the unit develops into a crystal with distinct faces and boundaries. When the crystal attains desired size and shape further growth could be stopped deliberately, fearing undesired growth and distortion. The harvested crystals could be used for device applications.

Salts of various monocarboxylic acids like formates and acetates and dicarboxylates like oxalates and malonates exhibit outstanding properties. Metal substituted acids like tartrates and lactates have interesting optical and ferroelectric characteristics. This study is about the growth, characterization and property evaluation of two metal coordination compound crystals, calcium malate and strontium
malate. This species of compounds are important mainly due to their non linear optical properties. The thesis encompasses nine main chapters describing various aspects associated with the experiment and analyses.

Depending upon the physical state of the starting material, crystallization techniques can be classified into melt growth, solid growth, vapour growth and solution growth. Growth of a crystal can also be achieved by the chemical reaction of two or more components. The first chapter discusses the various techniques employed for the growth of different types of crystals, giving emphasis to solution growth in general and gel growth in particular. Crystal growth from gel serves as an excellent alternative method when growth of monocrystals by conventional techniques causes problems due to decomposition before melting, non-availability of suitable flux, difficulty to control the temperature or lack of proper solvents. It is a simple, elegant method to grow single crystals under controlled growth at room temperature. A systematic study of crystallization in gel began in 1896 with Liesegang’s periodic crystallization experiments in silver dichromate. The main advantage of this method is its simplicity and effectiveness in growing single crystals. Crystals free from thermal strains can be developed by this method. It is an alternative to solution growth with controlled diffusion and the growth process is free from convection. So far, many carboxylate crystals such as tartrates, oxalates and amino acid salts like L-arginine phosphate and L-alanine phosphate that exhibit non liner optical properties and complexes of rare earth metals having remarkable optical and magnetic properties have been grown by the ionic diffusion process in gels.

Evaluation of the physical and chemical characteristics of the crystals is essential in finding their suitability for device grade applications. With the advancement of technology, a variety of analytical tools are available to the researchers which would help them inspect even the minute properties of the crystal. Spectroscopic characterization techniques like FTIR and FT Raman, single and powder X-ray diffraction techniques, thermogravimetric and differential thermal methods, vibration sample magnetometry, dielectric and microhardness techniques are employed in the study. The instrumentation details and the sample requirements for these analyses are discussed in the second chapter.
The development of defect free crystals of well defined morphology is of key interest to a crystal grower. In chapter 3, the growth procedure for calcium bis malate dihydrate crystals and strontium malate tetrahydrate crystals by ionic diffusion technique in single tubes is reported. There are various factors that affect the growth process and quality of a crystal. Various growth parameters like concentration of both the inner and outer reactants, pH of the growth medium and age of the gel must be controlled with utmost care. Optimum conditions for the growth of defect free crystals have been identified after careful investigation.

Structural investigation of a crystal is quite important in characterizing the material. X-ray diffractometry is the most reliable and widely used technique for this purpose. The diffraction techniques can be of two types namely, single crystal and powder X-ray diffraction methods. X-ray powder diffraction is used in the identification of crystalline phases in unknown samples, finding the lattice parameter and other lattice related information, quantitative determination of phases in multiphase samples, finding the orientation in single crystals, and stereographic projections. Single crystal x-ray diffraction is performed by analyzing the diffraction of X-rays from an ordered array of many identical molecules. Understanding the unit cell of these arrays simplifies the understanding of a crystal as a whole. Both single crystal and powder X-ray diffraction method have been employed for structural elucidation of the grown crystals. Crystal systems and symmetry groups of the title compounds have been identified for the first time and the details are presented in chapter 4.

Spectral characterization stands as a significant tool in recognizing the functional groups present in an unknown molecule and thus identifying it. Vibrational spectra of samples are recorded by both IR and Raman techniques. IR spectrum arises due to the absorption of light by vibrating molecules whereas Raman spectrum is due to the scattering of light by vibrating molecules. IR spectroscopy is an important non-destructive characterizing tool, which provides qualitative information regarding composition and structure of molecules under investigation. This may be of great use in predicting the structure of newly developed materials. The presence of a permanent dipole moment of a molecule may be regarded as a criterion for infra red spectra. Raman spectroscopy provides one with information regarding IR inactive molecules.
The characteristic vibrational frequencies would help identify the molecular species. For vibrations to be Raman active, it should produce a change in the molecular polarizability too. Analysis of the FTIR and FT Raman spectra of grown crystals forms the subject matter of the fifth chapter. Characteristic frequencies and corresponding functional groups are identified from the spectra.

Thermal investigations of a grown sample have an imperative role in its property evaluation. Details regarding the thermal stability, transition temperatures, and phase transitions can be obtained from the thermal treatment and thus help the quality control of materials. They find widespread use for both quality control and research applications on industrial products like polymers, metals and alloys. In the present case the thermal characterization of the grown crystals are done employing TG and DTA techniques. Chapter 6 deals with the analysis of the thermograms of calcium and strontium malate crystals. It gives information about the thermal stability and decomposition pattern of the compounds. The thermal studies give additional support to the composition and structure of the grown crystals as revealed by FTIR and X-ray analyses.

The dielectric and magnetic behaviour of materials help the technologist find their application for the appropriate device fabrication. Dielectric permittivity is dependent on parameters like temperature, orientation, grain size, molecular structure of the material and frequency of the applied field. The variation of dielectric constant and dissipation factor with frequency and temperature have been studied. The vibrating sample magnetometer is one of the standard equipment for the magnetic characterization of thin magnetic films and structures. It gives the absolute measure of the sample magnetization as a function of applied magnetic field and temperature. Magnetic susceptibility as well as the magnetic nature of the materials is investigated in the light of data obtained on VSM studies. The electrical and magnetic behaviour of the materials explored using above techniques are presented in chapter 7.

The suitability of a crystal for a particular technological application depends greatly on its mechanical strength. One of the highly reliable mechanical testing methods is the hardness measurement. Various other mechanical properties of the materials such as toughness, brittleness, yield strength, etc have specific correlation with hardness. The grown crystals are indent for different loads using a Clemex -
Microhardness Tester. The variation of microhardness with applied load was studied. Also, the relation between diagonal length and load was examined. All these information are presented in chapter 8.

The final chapter of the thesis is a concluding note which gives stress on the scope of the materials for future research and technology development. A comparison is made with other crystals of the same kind which were grown earlier.

The author has published some of the major portions of this work in refereed inter-national journals and some others have been communicated. Also, some of the works have been presented in different national seminars. The author has got a handful of other publications in closely related areas too. A list of these publications is appended.