

CHAPTER 1

INTRODUCTION TO NONLINEAR OPTICS AND CRYSTAL GROWTH

1.1 OVERVIEW ON NONLINEAR OPTICS

Nonlinear optics (NLO) is a new frontier of science and technology playing a major role in the emerging era of photonics. Photonics involves the application of photons for information and image processing and is branded to be the technology of the 21st century wherein nonlinear optical processes have applications in the vital functions such as frequency shifting, optical modulation, optical logic gates, optical switching and optical memory (He & Liu 1999). The invention of the first laser in 1960 created great interest on examining the interaction of optical materials with the light at higher intensities. Nonlinear optics is the study of the interaction of intense electromagnetic fields with materials to produce modified fields that are different from the input field in phase, frequency, amplitude and polarization. The light propagated through crystalline solid, which lacks a centre of symmetry, generates light at second and higher harmonics of the applied frequency (Chemla & Zyss 1987), (Sioncke et al 2003).

NLO materials typically have a distinct crystal structure which is anisotropic to electromagnetic radiation. Light passing through them undergoes a change in wavelength thereby releasing a photon of accumulated frequency. NLO materials can be used to double or triple the frequency of the laser light and are of considerable interest for the high speed processing of the



data which is essential for optical computing and optical telecommunication systems. The enhancement of the efficiency of the nonlinear process by the appropriate choice of NLO material constitutes a central concern in all these studies. The search for suitable materials exhibiting excellent second order nonlinear optical properties has been the focus of current research activity owing to their potential applications in optoelectronics, telecommunications and optical storage devices ‘Bella (2001)’, (Marder et al 1991), ‘Wang et al (1999)’, ‘(Cole et al 2000)’, (Zaitseva & Carman 2001).

The explanation of nonlinear effects lies in the way in which a beam of light propagates through a solid. The nuclei and associated electrons of the atoms in the solid constitute electric dipoles. The electromagnetic radiation interacts with these dipoles causing them to oscillate which obeys the classical laws of electromagnetism, results in the dipoles themselves acting as sources of electromagnetic radiation. If the amplitude of vibration is small, the dipoles emit radiation of the same frequency as the incident radiation. As the intensity of the incident radiation increases, the relationship between irradiance and amplitude of vibration becomes nonlinear resulting in the generation of harmonics in the frequency of radiation emitted by the oscillating dipoles. Thus frequency doubling or second harmonic generation (SHG) and indeed higher order frequency effects occur as the incident intensity is varied.

When an external field is applied, the material is polarized and constitutes a dipole moment within the medium since each constituent molecule acts as a dipole with a dipole moment P_i . The dipole moment vector per unit volume P is given by

$$P = \sum_i P_i \quad (1.1)$$



where the summation is over the dipole depends both on the properties of the medium and on the field strength. Thus one can write

$$P = \epsilon_0 \chi^{(1)} E \quad (1.2)$$

where ϵ_0 is the permittivity of free space. When the applied electric field strength is high enough and comparable to the inter-atomic field, the amplitude of the dipoles does not faithfully reproduce the exact sinusoidal electric field. Indeed, it produces the distorted reradiated wave which contains the induced polarization given by

$$P = \epsilon_0 \chi^{(1)} \cdot E + \chi^{(2)} \cdot E \cdot E + \chi^{(3)} \cdot E \cdot E \cdot E + \dots \quad (1.3)$$

where $\chi^{(1)}$ is the linear term which is responsible for the linear optical properties such as absorption, index of refraction, dispersion and birefringence of the medium. $\chi^{(2)}$, $\chi^{(3)}$ are the second and third order nonlinear susceptibilities and their magnitude decreases as their order increases (10^{-8} : 10^{-16} and so on). The second order susceptibility term $\chi^{(2)}$ gives rise to second harmonic generation, frequency mixing and parametric generation, while the third order susceptibility $\chi^{(3)}$ is responsible for the third harmonic generation, stimulated Raman scattering, optical bistability and phase conjugation. Some nonlinear optical phenomena are given in Figure 1.1(a) – (f).

For a material to be Nonlinear, it should have the following characteristics.

- (i) Large nonlinear figure of merit for frequency conversion
- (ii) High laser damage threshold



(iii) Fast optical response time

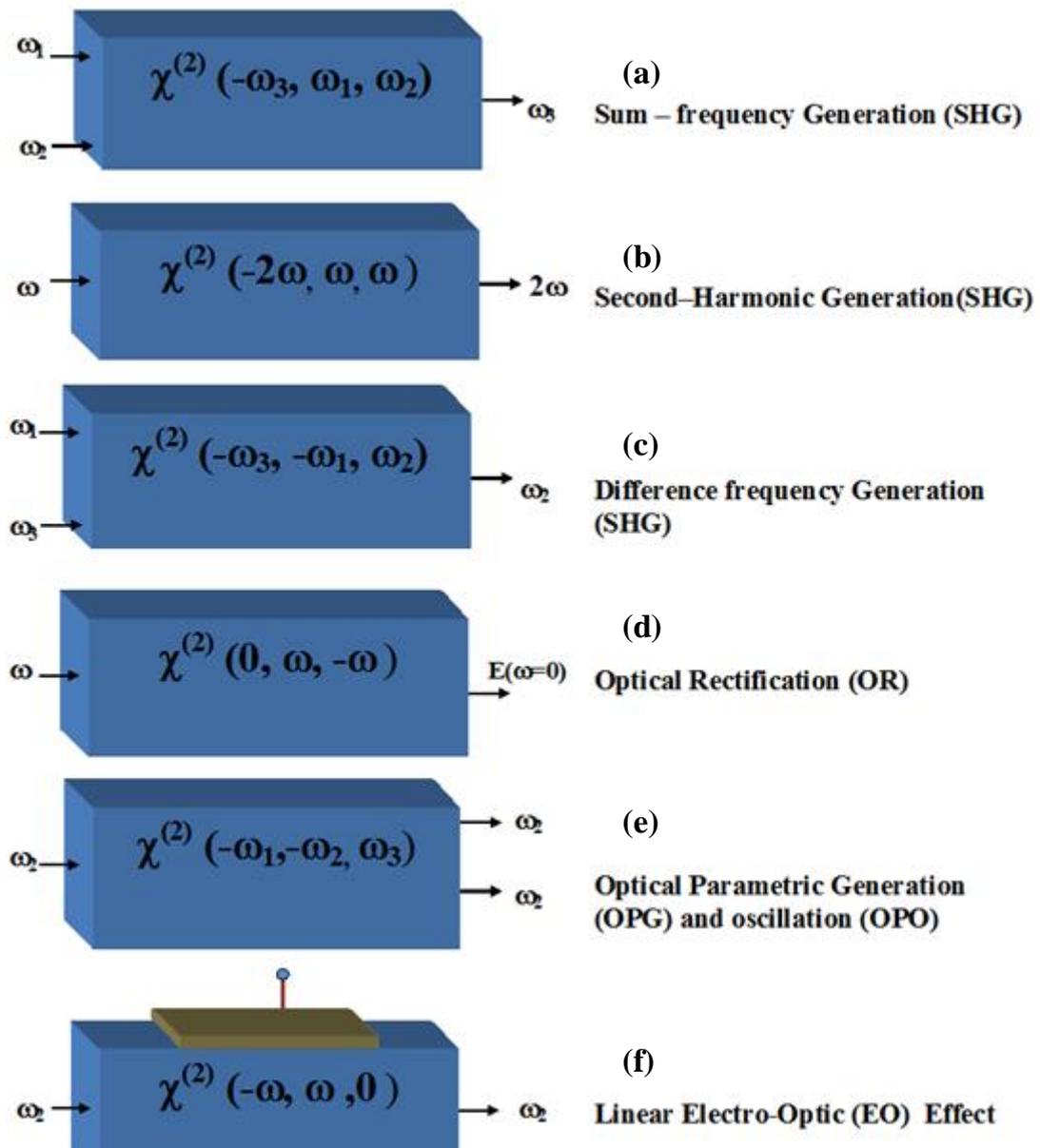


Figure 1.1 Nonlinear optical effects

(iv) Wide phase matchable range

- (v) Architectural flexibility for molecular design and morphology
- (vi) Ease of fabrication
- (vii) Non toxicity and environmental stability
- (viii) Optical transparency
- (ix) High mechanical strength and thermal stability.

1.1.1 Inorganic Crystals

These are covalent and ionic bulk materials where the optical nonlinearity is a bulk effect. The phenomenon of SHG in inorganic materials was first reported in 1961. The examples of these types of crystals are Lithium Niobate (LiNbO_3), Potassium Niobate (KNbO_3), Barium Titanate (BaTiO_3), Potassium Titanyl Phosphate (KTiOPO_4 , KTP), Potassium Dihydrogen Phosphate (KH_2PO_4 , KDP), Potassium Deuterium Phosphate (KD^*P), Ammonium Dihydrogen Phosphate (NH_3PO_4 , ADP), Lithium Iodate (LiIO_3), etc. Many of these materials have been successfully used in commercial frequency doublers, mixers and parametric generators to provide coherent laser radiation with high frequency conversion efficiency in the new region of the spectrum inaccessible by other nonlinear crystal conventional sources. Lithium niobate (LiNbO_3) crystals are one of the most investigated materials for widespread and promising applications in nonlinear optics, e.g., for parametric amplification and second-harmonic generation, holographic data storage, optical information processing, phase conjugation and wavelength filters (Bolt & Bennema 1990). The inorganic NLO materials have some advantages like high melting point, high mechanical strength and compatible physical properties, on the other hand, such materials suffer from disadvantages like modest optical nonlinearity due to the lack of extended



π -electron dislocation, absorption in the visible region, poor response time and degradative photorefractive effects, low laser damage threshold ($\sim 10 \text{ MWcm}^{-2}$) and poor optical transparency (Hussaini et al 2009).

1.1.2 Organic Crystals

In the recent past, the extensive investigations are carried out on organic materials due to their high nonlinearity compared to inorganic materials. The origin of nonlinearity in the NLO material is due to the presence of delocalized π -electron system connecting donor and acceptor groups which enhance the asymmetric polarization. Recently, an extremely large number of organic compounds with non-localized π -electron systems and a large dipole moment have been synthesized to realize the nonlinear susceptibilities far larger than the inorganic optical materials (Chemla & Zyss 1987). Important organic materials that have been studied by many researchers are: L-threonine, L-threonine formate, L-alanine, L-phenylalanine, L-arginine L-lysine monohydrochloride dihydrate (L-LMHCl), nitroaniline, L-alaninium maleate etc. (Hanumantharao & Kalainathan 2012), (Prakash et al 2011), (Senthil et al 2009). Despite of high nonlinearity, the applications of organic NLO crystals have developed rather slowly because the organic molecules are constructed by weak van der Waal forces and hydrogen bonds, and it is difficult to grow large optical quality single crystals and also, due to often fragile nature of these crystals. The shortcoming of these crystals, such as poor physico- chemical stability, low hardness and cleavage tendency and poor thermal strength obstruct their device applications.

Materials with nonlinear electro-optic properties have a role in modern opto-electronics that is analogous to that of nonlinear electronic circuit elements in conventional electronics. Typical function of such element is to modulate carrier waves amplify and rectify signals and acts as very fast



switches. In amorphous solids, liquids and garnets in solids in which the molecules are related by a centre of symmetry, the even terms are zero.

The two particular nonlinear effects, the linear electro-optic effect or Pockels effect through the refractive index of the material varies with the applied electric field and second harmonic generation where irradiation of the solid with light of one frequency results with the light of double the frequency.

1.1.3 Semiorganic Materials

In order to retain high NLO efficiency of organic materials and overcome the shortcoming, a new strategy of synthesizing organic–inorganic hybrid compounds have been proposed, thus leading to a new class of materials namely, the semiorganics. The relatively strong ligand bond permits the complex crystals to combine the advantages of inorganic crystals, such as good stability, with the advantages of organic crystals, such as high nonlinear and molecular engineering features. In recent years, semiorganic crystals find considerable interest among researchers due to their high damage threshold, wide transparency range, less deliquescence, excellent nonlinear optical coefficient, low angular sensitivity and exceptional mechanical properties (Xing et al 1987), ‘Velsko (1990)’, (Gupte et al 2002), (Rajesh et al 2004).

1.2 CRYSTAL GROWTH

Present day advanced technologies heavily rely on precisely characterized high quality crystals. Crystals find applications in electronics, lasers, light valves, laser communication devices ‘Carmino Salvo (1971)’, light emitting diodes, thermal imaging, pyroelectric detectors (Satapathy et al 2002), X-ray spectroscopy (Srinivasan et al 1999), inertial confinement fusion (Zaitseva et al 1999), etc. Large size crystals are usually essential for



device fabrication, and it must be possible to grow crystals of good optical quality for utilization (Tang 1995).

Efforts are made to grow large crystals in short durations by fast-growth techniques. Crystal growth is basically a process of arranging atoms, ions, molecules or molecular assemblies into regular three-dimensional periodic arrays. Crystal growth can be treated as an important branch of materials science leading to the formation of technologically important materials of different sizes. When a crystal is in dynamic equilibrium with its mother phase, the free energy is minimum and no growth can occur. This equilibrium has to be disturbed suitably for growth to occur. This may be done by an appropriate change in temperature, pressure, pH, chemical potential, etc. The driving force for crystallization actually derives from supersaturation, supercooling of liquid or gas phase with respect to the component whose growth is required. Nucleation or crystallization center is an important feature of crystal growth. Nucleation may occur either spontaneously due to the conditions prevailing in the parent phase or it may be induced artificially. In case of organic crystals, delocalized π -electrons can easily move between electron donor and electron acceptor groups on opposite sides of the molecule, inducing a molecular charge transfer (Gallagher et al 2003), (Chemla & Zyss et al 1987), (Bosshard et al 1993).

Growth of crystal ranges from a small inexpensive techniques to a complex sophisticated expensive process and the crystallization time ranges from minutes, hours, days and to months (Brice 1986). The choice of a particular method depends on the material to be crystallized, its quality, size, growth rate and in particular the physico-chemical properties of the material. Growth from liquid is used for the present investigation. Crystal growth is a diffusion process; solute molecules or ions reach the growing faces of a crystal by diffusion through the liquid phase. On reaching the surface, the



molecules or ions must be accepted by the crystal and organized into the space lattice. The reaction occurs at the surface at a finite rate and the overall process consists of two steps in series. Neither the diffusion nor the interfacial step will proceed unless the solution is supersaturated. The crystal growth process involves phase transitions of the type, solid-solid, liquid-solid and vapour-solid. The crystallization process consists of three steps.

- (i) Achievement of supersaturation or supercooling
- (ii) Formation of crystal nuclei
- (iii) Growth of crystals

All the above processes may occur simultaneously at different regions of a crystallization unit. However an ideal crystallization process consists of a strictly controlled stepwise program.

1.3 CRYSTAL GROWTH TECHNIQUES

The process of crystal growth is controlled by change of state, or phase change, to the solid state. This transition may occur from the solid, liquid or vapor state. Depending on the phase transitions involved in the process, the crystal growth methods can generally be classified into four main categories (Sankaranarayanan & Ramasamy 2005), 'Sankaranarayanan (2005)' and Brice (1965).

- (i) Solid growth (solid to solid)
- (ii) Vapor growth (vapor to solid)
- (iii) Melt growth (liquid to solid)
- (iv) Solution growth (liquid to solid)



The conversion of a polycrystalline piece of material into single crystal by causing the grain boundaries to be swept through and pushed out of the crystal takes place in the solid-growth of crystals 'Mullin (1972)'. Solid growth processes are rarely used except for certain metals where strain annealing is effective and certain cases where a crystal structure change occurs between the melting point and room temperature. An efficient process is the one, which produces crystals adequate for their use at minimum cost. Better choice of the growth method is essential because it suggests the possible impurity and other defect concentrations. Choosing the best method to grow a given material depends on material characteristics. It seems that the essential task for the crystal growers at present is to gain basic knowledge about the correlation of given material with crystal growing method and the growth conditions defined to be special parameters. On analyzing these points one can find which technique is appropriate for a given material. Various growth techniques are discussed in proceeding sections with more emphasis on solution growth technique.

1.3.1 Growth from Solids

In this method, by the application of internal strain to the amorphous solid material, the constituent atoms or molecules may rearrange in a crystalline form 'Goodman (1974)'. Solid growth processes are rarely used except for certain metals where strain annealing is effective and certain cases where a crystal structure change occurs between the melting point and room temperature.

1.3.2 Vapor Growth

Vapor growth techniques can be adopted for the growth of materials which lack a suitable solvent and sublime before melting at normal pressure. Vapor growth methods have been employed to produce bulk crystals



and to prepare thin layers on crystals with a high degree of purity (Faktor & Garrett 1974). Crystals of high purity can be grown from vapor phase by sublimation, condensation and sputtering of elemental materials 'Brenner (1963)', 'Reynolds (1963)'. To obtain single crystals of high melting point materials this method is used. Growth from vapor phase may generally be subdivided into:

- (i) Physical vapor transport
- (ii) Chemical vapor transport.

There is an increasing demand for vapor grown crystals because these crystals sometimes possess unusual properties. The crystals often have perfectly flat external faces that cannot be duplicated by other means and they often contain less substructure and imperfections than melt grown crystals. The fundamental aspects of vapor phase crystal growth might be divided into four major areas:

- (i) Thermodynamics, which controls the driving force for the chemical reaction occurring at the vapor solid interface.
- (ii) Mass transport, by which the reactants reach the growing surface and products are removed.
- (iii) Surface kinetics including adsorption of reactants onto the surface, surface diffusion, step generation, surface chemical reactions between adsorbed reactants at steps and desorption of products.
- (iv) Structural aspects of growth, especially epitaxial growth such as defect generation and surface morphology of homo epitaxial and hetero epitaxial layers.



1.3.3 Melt Growth

Crystal growth from melt is the most widely practiced commercial process for the production of single crystals. Melt growth is the process of crystallization by fusion and resolidification of the pure material. A crystal can be grown by this method only if it melts congruently, i.e., the substance should not decompose before melting and undergo phase transition at temperatures below the melting point of the substance. In this technique apart from possible contamination from crucible material and surrounding atmosphere, no impurities are introduced in the growth process and the rate of growth is normally much higher than the other methods (Sankaranarayanan & Ramasamy 1998). The growth from melt can be classified as follows

With crucibles

- (i) Bridgman -Stockbarger technique
- (ii) Czochralski technique
- (iii) Kyropoulos technique
- (iv) Zone melting technique

Without crucibles

- (i) Floating zone technique
- (ii) Verneuil technique

1.3.3.1 Bridgman -Stockbarger technique

This technique was named after its inventor 'Bridgman (1924)'. In this process the material to be grown is taken in a vertical cylindrical container, tapered conically with a point bottom and made to melt using a



suitable furnace. The container is lowered slowly from the hot zone of the furnace into the cold zone. The rates of movement mostly taken in the range 1-30 mm/hr. Crystallization begins in the tip and continues usually by growth from the first formed nucleus (Scheel & Fukuda 2004), (Bardsley et al 1977), 'Brice (1965)', 'Bridgman (1924)'. The relative advantages of this technique are low cost, with added advantage that the melt temperature increases with distance from the solid-liquid interface and the system is therefore density stable.

The Bridgman technique cannot be used for materials which decompose before melting or which undergo solid state phase transformation between their melting points and the temperature to which they will be cooled. A complete Bridgman setup is shown in Figure 1.2.

A disadvantage is that the melt is in contact with the container for longer periods, increasing the risk of impurity pick-up. The second disadvantage is that adhesion of the solid materials to the ampoule wall or compression of the solid by the contracting container during cooling can lead to the development of stresses high enough to nucleate dislocations in the material. A further drawback of the technique is that the container wall acts as a preferential, spurious nucleation site, resulting in polycrystalline rather than single crystal growth unless the temperature gradient and liquid - solid interface shape are well controlled.



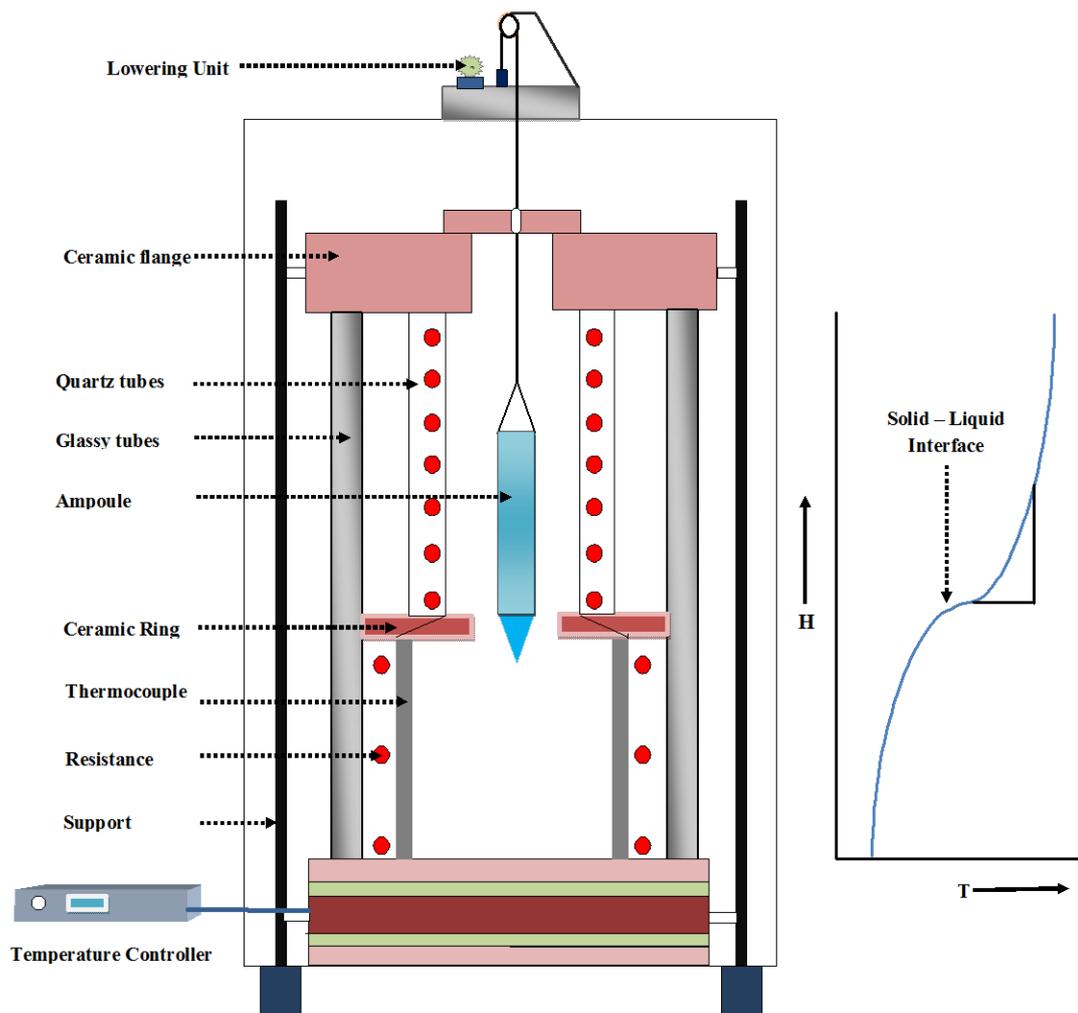


Figure 1.2 Schematic of Bridgman technique

1.3.3.2 Czochralski technique

Czochralski technique is a popular method of crystal growth because it can produce large, dislocation free crystals in a relatively short space of time 'Brice (1965)'. The Czochralski growth geometry is shown schematically in Figure 1.3. The material to be grown is melted by induction or resistance heating under a controlled atmosphere in a suitable non-reacting container (crucible). By controlling the furnace temperature, the material is melted. The melt temperature is then adjusted to be slightly above the melting point and seed crystal is lowered. After thermal equilibrium, the seed is

contacted with the melt and the melt temperature is raised to establish the desired growth interface configuration. The seed is rotated at the same time in order to attain thermal symmetry and also to stir the melt. When the temperature of the seed is maintained very low compared to the temperature of the melt, by suitable water cooling arrangement, the molten charge in contact with the seed will solidify on the seed. The advantage of this method over the Bridgman method is that it can accommodate the volume expansion associated with the solidification.

The main requirements for growing good crystals by this method are pulling and rotation rates should be smooth and that the temperature of the melt should be accurately controlled. The diameter of the crystal is dependent on both the temperature of the melt and the pull rate. To increase the diameter of the crystal being grown, the power is reduced or the pull rate decreased, the opposite applies for reducing the diameter. The maximum size of crystal that can be produced by this technique is governed by the volume of the melt, the diameter of the crucible, the distance that the pull rod can move and the strength of the crystal at its minimum diameter. It is essential for this method that the temperature distribution in the melt should be such that the temperature of the solid-liquid interface is the lowest in the liquid. If it is not, spurious nucleation will occur. Czochralski method has gained wide recognition particularly in growing single crystals of semiconductors like silicon and other materials. Liquid Encapsulated Czochralski abbreviated as LEC technique makes it possible to grow single crystals of materials, which consists of components that produce high vapour pressure at the melting point. This refined method of Czochralski technique is widely adopted to grow III-V compound semiconductors.



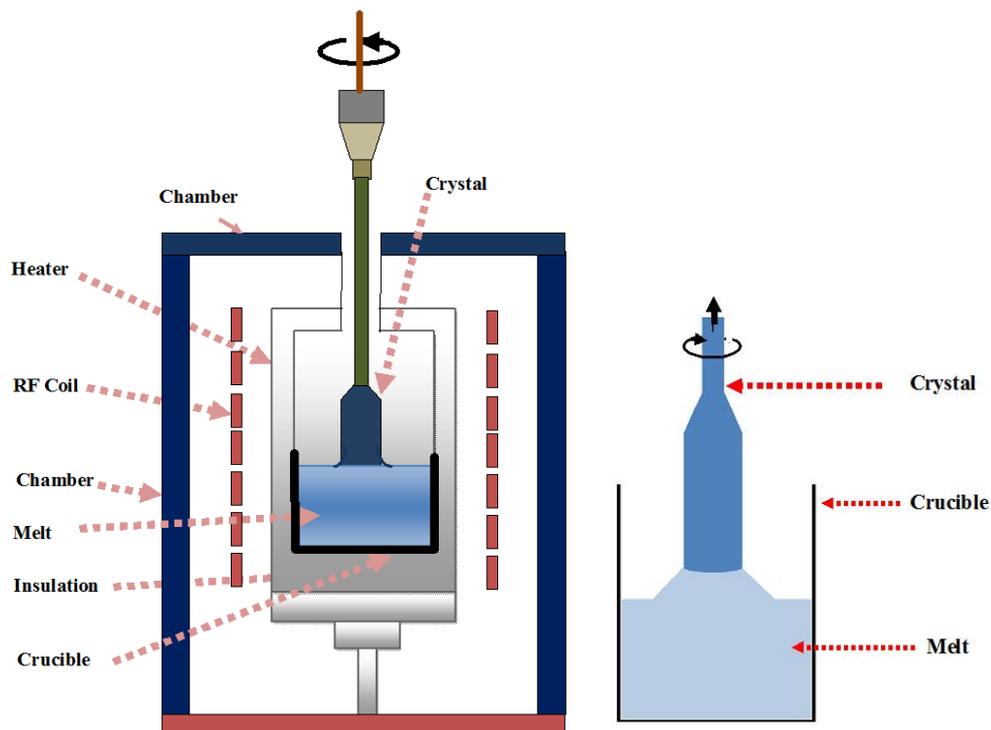


Figure 1.3 Schematic of Czochralski technique

1.3.4 Solution Growth

Solution growth can be adopted to grow single crystals of those materials that are highly unstable in the melt and undergo decomposition upon melting. Advantages of this technique are convenience, simplicity and possible avoidance of complex growth apparatus. This method envisages that the material must crystallize from solution with prismatic morphology.

The basic growth procedure involves seeded growth from a saturated solution that regulates its growth by the control of temperature and stirring of the solution for homogeneous concentration and temperature gradient. Crystal growth is in need of production of an exact supersaturated solution at the growing face. This supersaturation is achieved either by lowering the temperature of the solution or by slow evaporation of the solvent

'Mullin (1976)'. Solution growth by itself can be classified into two major types

- (i) Low temperature solution growth
- (ii) High temperature solution growth

1.3.4.1 Low temperature solution growth

The choice of the solvent is probably the most critical step in low temperature solution growth. For many materials, water is found to be a good solvent. It is however fairly volatile which can be overcome by adding a film of oil on the surface. For some materials, it is necessary to use mixed solvents to reduce the solubility. The solution used is frequently compatible with a wide range of glasses and plastics. This method requires precise temperature control. The advantages of crystal growth from low temperature solution near the ambient temperature results in simple equipment design, which gives a good degree of control to an accuracy of ± 0.01 °C.

The growing crystals are completely surrounded by the solution and growth occurs on many flat faces. This makes the stirring of solutions much more complicated. But vigorous stirring can lead to cavitations or turbulence, which may result in spurious nucleation. Spurious nucleation is encouraged by temperature irregularities and foreign particles in the solution. In order to prevent dissolution of the seed or rapid initial growth, the solution must be almost exactly saturated at the initial growth temperature before the seed crystal is inserted. A common technique is to make a slightly undersaturated solution and to allow evaporation to bring it to saturation. Seed crystals should be prepared with care. It should be noted that the surface of the seed crystal should not have been damaged 'Scheel (1994)'.



Low temperature solution growth can be subdivided into three methods

- (i) Slow cooling method
- (ii) Slow evaporation method
- (iii) Temperature gradient method

1.3.4.2 Slow cooling method

It is the easiest way to grow crystals by solution growth technique. Its main disadvantage is the need to use a range of temperature. The possible range is usually small so that much of the solute remains in the solution at the end of a run. To compensate this effect, large volumes of solutions are required. The use of a range of temperature may not be desirable because the properties of the growth material may vary with temperature. Eventhough the method has technical difficulty of requiring a programmable temperature control; it is widely used with great success.

1.3.4.3 Slow evaporation method

This method is similar to the slow cooling method in view of the apparatus requirements. The temperature is fixed constant and provision is made for evaporation. With non-toxic solvent viz. water; it is permissible to allow evaporation into the atmosphere. Typical growth conditions involve temperature stabilization to about ± 0.005 °C and rate of evaporation of a few mm^3/h . The evaporation techniques of crystal growth have the advantage that the crystals grow at a fixed temperature. But inadequacies of the temperature control system still have a major effect on the growth rate. This method can be adopted for materials, which have very small temperature coefficient of solubility.



1.3.4.4 Temperature gradient method

This method involves the transport of materials from a hot region containing the source material to be grown to a cooler region where the solution is supersaturated and the crystal grows. The main advantages of this method are

- i. The crystals grow at a fixed temperature
- ii. This method is insensitive to changes in temperature provided both the source and the growing crystal undergo the same change
- iii. Economy of solvent

On the other hand, changes in the small temperature difference between the source and the crystal have a large effect on the growth rate. Earlier there were crystallizers in which a single chamber is used with a bottom heater. Nutrient is placed at the bottom of the bath and the slightly hotter solution is pumped from there up the spider support and then allowed to flow downward over the crystals.

1.3.5 Unidirectional Growth Method from Solution

To grow high quality crystal with a reasonable yield, the investigation of a possible single crystal growth technique is needed. Nonlinear optical interactions in bulk single crystals usually require phase-matching orientations in acentric crystals to maximize the frequency conversion efficiency. To attain high wavelength conversion efficiency, nonlinear optical crystals have to be long enough in the phase-matched direction. Therefore, method of growing nonlinear optical crystals with a large size in a phase-matched direction is strongly required. Further, growth



of organic or inorganic crystal with specific orientation has tremendous value in terms of its significance towards device application. From this point of view, a novel Sankaranarayanan-Ramasamy crystal growth method has more advantages when compared to conventional solution growth techniques (Sankaranarayanan & Ramasamy 2005) and this can be employed to grow unidirectional crystal from solution.

The main concept of the method is gravity driven concentration gradient. The solutions at the bottom of the ampoule have more concentration compared to top solutions. The concentration gradient is directly proportional to time.

1.3.5.1 Sankaranarayanan-Ramasamy (SR) method

It is one of the methods to grow the crystals from solution. Unidirectional crystals are very important for the preparation of functional crystals. For example, as the conversion efficiency of second harmonic generation is always highest along the phase-match direction for nonlinear optical crystals, the unidirectional crystal growth method is most suitable for the crystal growth along that direction. In addition, the unidirectional solution crystallization usually occurs at around room temperature; much lower thermal stress is expected in these crystals over those grown at high temperatures. This is particularly helpful for growth of mixed crystals because thermal stresses can cause these crystals to crack easily. Crystals with all facets and different morphology are grown by conventional solution growth technique but from application point of view, orientation controlled good quality, large size SHG crystals are needed. In all the methods of growth by solution, planar habit faces contain separate regions common to each facet having their own sharply defined growth direction known as growth sectors. The boundaries between these growth sectors are more strained than the extended growth sectors due to mismatch of lattices on either side of the



boundary as a result of preferential incorporation of impurities into the lateral section (Gallagher et al 2003).

In Sankaranarayanan-Ramasamy (SR) method, a glass ampoule was made up of an ordinary hollow borosilicate-glass with a tapered V-shaped or flat bottom portion to mount the seed crystal and a U-shaped top portion to fill a good amount of saturated solution to grow a good size crystal. The middle portion was cylindrical in shape with lesser diameter than that of the U-shaped portion, wherein one can get a cylindrical shaped crystal. The schematic of experimental set up of Sankaranarayanan – Ramasamy method is shown in Figure 1.4.

1.3.5.2 Salient features of Sankaranarayanan-Ramasamy (SR) method

The salient features of SR method are listed below:

- (i) Single crystal with desired orientation is possible at room temperature.
- (ii) It is easy to adjust the growth rate as per our need.
- (iii) Scaling up is relatively very simple.
- (iv) With a thin plate as seed, growth of large size crystal is possible.
- (v) Microbial growth has been causing serious concern in solution growth experiments largely due to aging of the solution. However, as fresh solution can be constantly fed as crystal growth proceeds the problem associated with microbial growth can be avoided.



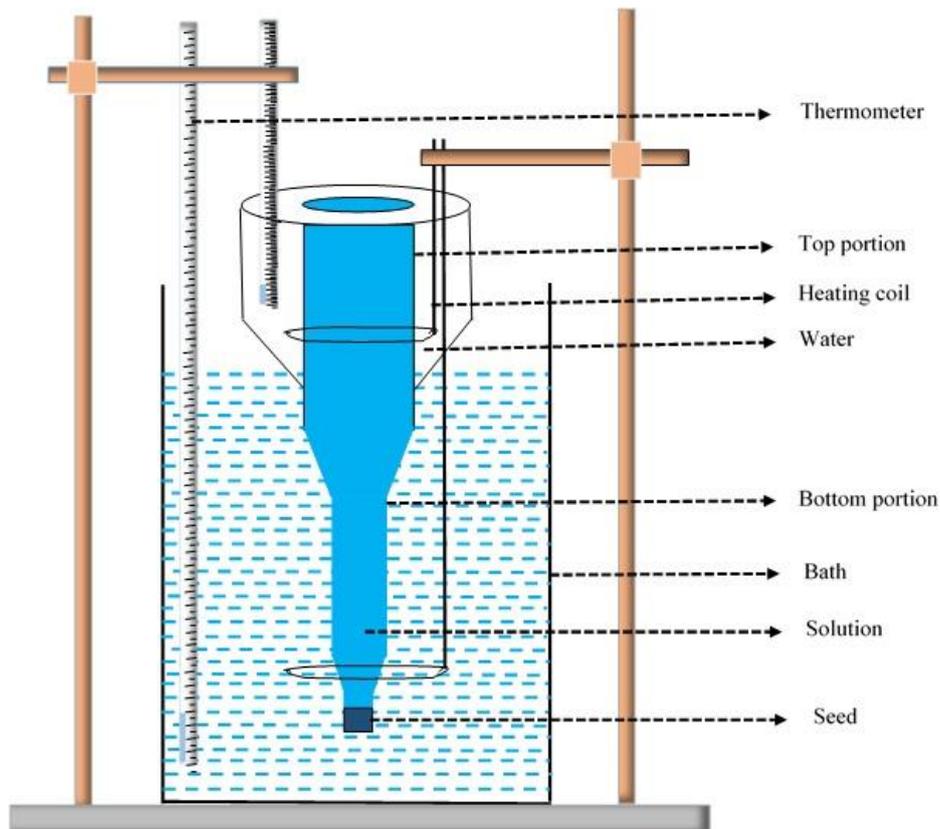


Figure 1.4 Schematic of experimental setup of Sankaranarayanan-Ramasamy method

- (vi) Simple experimental set up offers the feeding of the growth solution at a definite interval which depends on the growth rate of the crystal, thereby minimizing the exposure of the growth solution to the environment.
- (vii) The achievement of solute-crystal conversion efficiency of 100% reduces the preparation and maintenance of growth solution to a large extent because in conventional solution growth method, to grow such a large size crystal, a large quantity of solution in a large container is normally used and only a small fraction of the solute is converted into bulk single crystal. But, in the present method, the size of the growth ampoule is the size of the crystal.