2. LITERATURE SURVEY

2.1 INTRODUCTION OF NONLINEAR OPTICS

This chapter analyzes the literature survey of the nonlinear optics in detail. The high speed and ease of production of photons (light), the area of photonics are the active fields of research in view of modern society’s demand and also for improved telecommunication, data storage, retrieving, processing and transmission. The design of devices and utilization of photons instead of electrons for the transmission of information has created a need for new materials with unique nonlinear optical (NLO) material [21]. The nonlinear optics is the field which includes all phenomena in which optical parameters of materials are changed with the interaction of intense coherent source of light. The nonlinear optical phenomenon leads to the enhancement of understanding light-matter interactions. The searches for new molecular materials with nonlinear optical properties are the subject of considerable importance investigations in their potential applications in photonic devices [22]. The invention of the laser has created a revolution in research and development in the area of photonics and the investigations on nonlinear optical properties enhanced and widened the horizon of application of lasers. The drawback of most of the laser materials is their inability to generate the radiation in a wide spectral region, such as for some applications the appropriate laser materials exist and thermal properties of source materials the emitted power is restricted. The nonlinear optics makes it possible to transfer the energy from wavelength to another and hence provide a solution for the radiation sources in the spectra range of radiation [23]. The nonlinear optical (NLO) single crystals therefore, could be employed to generate the sources of different wavelengths for which the lasers are not available. The output radiation beam from a nonlinear optical device is the similar properties of a laser source that could be directly employed as laser source. For certain applications highly powerful laser radiation is used in inertial confinement fusion research [24]. The field of nonlinear optics emerged nearly five decades ago with the development of the first operating laser and the demonstration of frequency doubling phenomena. These milestone
discoveries not only created much interest in laser science, but also set the scope for future research work in nonlinear optics. The extraordinary growth and development of nonlinear optical materials during the past decade had rendered photonic technologies, an indispensable part of daily life. With the emerging demand for information systems, nonlinear optical materials are considered as the key elements for the future photonic technologies of optical computing, telecommunications, optical interconnects, high density data storage, sensors, image processing, switching etc. The second harmonic generation (SHG) is a nonlinear optical process that results in the conversion of an input optical wave into an output, twice the input frequency. The process occurs within a nonlinear medium, usually a crystal. The light propagated through a crystalline solid, which lacks a centre of symmetry, generates light at second and higher harmonics of the applied frequency. Such frequency doubling processes are commonly used to produce green light (532 nm), for example, an Nd: YAG (Neodymium yttrium-aluminium-garnet) laser operating at 1064 nm. This important nonlinear property of non-centro symmetric crystals is called second harmonic generation (SHG) and this phenomenon and the materials in which it occurs are the subjects of intense study [25].

2.2 THEORETICAL EXPLANATION OF NONLINEAR OPTICS

The explanation of the nonlinear effect lies in the way in which a beam of light propagates through solid, nuclei and associated electrons of the atoms in the solid form the electric dipoles.

The electromagnetic radiation interacts with the dipoles causing them to oscillate, the classical laws of electromagnetism results in the dipoles that acts as sources of electromagnetic radiation. The phase velocity and wavelength of this electromagnetic wave are determined by $n_2$, the refractive index of the doubled frequency. To obtain high conversion efficiency, the phase vectors of input beams and generated beams are to be matched. 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. The frequency doubling or second harmonic generation (SHG) and indeed higher order frequency effects occur as the incident, intensity is increased. In a nonlinear medium, the induced polarization is a nonlinear function of the applied field. A medium exhibiting second harmonic generation (SHG) is a crystal composed of molecules with asymmetric charge distributions arranged in the crystal in such a way that a polar orientation is maintained throughout the crystal.

At very low fields, the induced polarization is directly proportional to the electric field [4].

\[ P = \varepsilon_0 \chi E \]  

(2.1)

where \( \chi \) is the linear susceptibility of the material, \( E \) is the electric field vector, \( \varepsilon_0 \) is the permittivity of free space. At high fields, the polarization becomes independent of the field and the susceptibility becomes field dependent. Therefore, this nonlinear response is expressed by writing the induced polarization as a power series in the field.

\[ P = \varepsilon_0 (\chi^{(1)} E + \chi^{(2)} E . E + \chi^{(3)} E . E . E + ...) \]  

(2.2)

In nonlinear terms, the product of two or more oscillating fields give oscillation at combination of frequencies and therefore, the above equation could be expressed in terms of frequency as:

\[ P(-\omega_0) = \varepsilon_0 \{ (\chi^{(1)}(-\omega_0;\omega_1) . E(\omega_0) + \chi^{(2)}(-\omega_0;\omega_1,\omega_2) . E\omega_1 . E\omega_2 \\
+ \chi^{(3)}(-\omega_0;\omega_1,\omega_2,\omega_3) . E\omega_1 . E\omega_2 . E\omega_3 + ...) \} \]  

(2.3)

where \( \chi^{(2)}, \chi^{(3)} \ldots \) are the nonlinear susceptibilities of the medium. \( \chi^{(1)} \) is the linear term responsible for material’s linear optical properties like, refractive index, dispersion, birefringence, and absorption. \( \chi^{(2)} \) is the quadratic term which describes second harmonic generation in non-centrosymmetric materials. \( \chi^{(3)} \) is the cubic term responsible for third harmonic generation, stimulated Raman scattering, phase conjugation and optical instability.
Hence, the induced polarization is capable of multiplying the fundamental frequency to second, third and even higher harmonics. The coefficients of \( \chi^{(2)}, \chi^{(3)} \) and \( \chi^{(3)} \) give rise to certain optical effects, which are listed in Table 2.1. If the molecule or crystal is centrosymmetric, then \( \chi^{(2)} = 0 \). If a field \( +E \) is applied to the molecule (or medium), equation 2.3 predicts that the polarization induced by the first nonlinear term is predicted to be \( +E^2 \). Yet if the medium is centrosymmetric the polarization should be \( -E^2 \). This contradiction could only be resolved if \( \chi^{(2)} = 0 \) in centrosymmetric media. If the same argument is used for the next higher order term, \(+E\) produces polarization \( +E^3 \) and \(-E\) produces \(-E^3\), so that \( \chi^{(3)} \) is the first non-zero nonlinear term in centrosymmetric media.

\[
2\omega_1 = \omega_2 \tag{2.4}
\]

During this process, a polarized wave at the second harmonic frequency \( 2\omega_1 \) is produced. The refractive index \( n_1 \) is defined by the phase velocity and wavelength of the medium. The energy of the polarized wave is transferred to the electromagnetic wave at a frequency \( \omega_2 \). The phase velocity and wavelength of this electromagnetic wave are determined by \( n_2 \), the refractive index of the material corresponding to the doubled frequency. To obtain high conversion efficiency, the phase vectors of input beams and generated beams are to be matched.

\[
\Delta K = \frac{2\pi}{\lambda(n_1 - n_2)} = 0 \tag{2.5}
\]

where \( \Delta K \) represents the phase-mismatch. The phase-matching could be obtained by angle tilting, temperature tuning or by other methods. Hence, to select a nonlinear optical crystal, for a frequency conversion process, the necessary criterion is to obtain high conversion efficiency. The conversion efficiency \( \eta \), is given by
Table 2.1 Optical effects of nonlinear materials

<table>
<thead>
<tr>
<th>Order</th>
<th>Susceptibility</th>
<th>Optical effects</th>
<th>Applications</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$\chi^{(1)}$</td>
<td>Refraction</td>
<td>Optical fibers</td>
</tr>
<tr>
<td>2</td>
<td>$\chi^{(2)}$</td>
<td>SHG ($\omega + \omega = 2\omega$)</td>
<td>Frequency doubling</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Frequency mixing</td>
<td>Optical parametric oscillators</td>
</tr>
<tr>
<td></td>
<td></td>
<td>($\omega_1 \pm \omega_2 = \omega_3$)</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>Pockel effect ($\omega + 0 = \omega$)</td>
<td>Electro-optic modulators</td>
</tr>
<tr>
<td>3</td>
<td>$\chi^{(3)}$</td>
<td>4 wave mixing Phase gratings</td>
<td>Raman coherent spectroscopy Real-time holography</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Kerr effect</td>
<td>Ultra high-speed optical gates</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Optical amplitude</td>
<td>Amplifiers, choppers etc.</td>
</tr>
</tbody>
</table>

\[ \eta = PL^2 \left( \frac{d_{\text{eff}} \sin \Delta K L}{\Delta K L} \right)^2 \]  \tag{2.6} 

where $d_{\text{eff}}$ is the effective nonlinear coefficient, $L$ is the crystal length, $P$ is the input power density and $\Delta K$ is the phase-mismatching. In general, higher power density, longer crystal, large nonlinear coefficients and smaller phase mismatching would result in higher conversion efficiency [26].

### 2.3 THE TYPES OF NONLINEAR OPTICAL EFFECTS

Some nonlinear optical processes are familiar to physicists, chemists, and other scientists because they are in common use in the laboratories. The second harmonic generation is a nonlinear optical process that results in the conversion of an input optical wave into an output wave of twice as that of the input frequency. The process occurs within a nonlinear medium, usually a
crystal (KDP-Potassium di hydrogen phosphate, KTP-Potassium Titanyl Phosphate, etc.). Such frequency doubling processes are commonly used to produce green light (532nm) using a Nd:YAG (Neodymium:Yttrium Aluminum Garnet) laser operating at 1064 nm [27].

Some of the NLO processes are given below:

(a) Second harmonic generation
(b) Sum frequency generation
(c) Difference frequency generation
(d) Optical parametric generation
(e) Linear electro-optic effect or Pockel’s effect
(f) Optical rectification

2.3.1 Second harmonic generation

The process transformation of light with frequency ‘ω’ into the light with double frequency 2ω and half the wavelength (Figure 2.1) are referred to second harmonic generation (SHG). The process is spontaneous and involves three photon transitions. A second harmonic generation has been of practical interest ever since after it was demonstrated because of its efficient conversion from fundamental to second harmonic frequencies. This could be achieved by the available powerful sources of coherent radiation at higher to attainable wavelengths [28].

Figure 2.1: Schematic diagram of SHG.
The most extensively studied conversion process of all has been the doubling of the 1.064 μm line obtained from the neodymium ion in various hosts. In particular, the doubling of the continuous wave Nd:YAG laser source is recently the subject of intensive study, because the laser light itself is efficient and powerful when the green light obtained by doubling is placed spectrally for detection by photomultipliers.

2.3.2 Sum frequency generation

It is a nonlinear optical process. Crystal materials with inversion symmetry could exhibit nonlinearity. In such NLO materials, the sum frequency generation could occur. Figure 2.2 illustrates the sum frequency generation.

\[ \omega_1 + \omega_2 = \omega_3 \] (2.7)

when two electromagnetic waves with the frequency \( \omega_1 \) and \( \omega_2 \) interact in an NLO medium, a nonlinear polarizability could be induced. The NLO material generates an optical wave of frequency \( \omega_3 \) which is equal to the sum of the two input wave frequency \( \omega_1 \) and \( \omega_2 \). The energy of output wave is represented by the equation 2.7.

Figure. 2.2: Schematic diagram of sum frequency generation.

2.3.3 Difference frequency generation

The process of difference-frequency generation is described by the following equation. Figure.2.3 illustrates the difference frequency generation. Here the frequency of the generated wave is the difference of those of the input frequencies.

\[ \omega_1 - \omega_2 = \omega_3 \] (2.8)
2.3.4 Optical parametric generation

The optical parametric generation (Figure. 2.4) is an inverse process of sum frequency generation. It splits one high-frequency photon (pumping wavelength $\lambda_p$) into two low-frequency photons (signal wavelength $\lambda_s$ and idler wavelength $\lambda_i$)

$$\omega_s - \omega_i = \omega_p \tag{2.9}$$

![Figure. 2.4: Schematic diagram of optical parametric generation $\omega_i$.](image)

2.3.5 Linear electro-optic effect

The Pockel's effect is a linear change in the refractive index of a medium in the presence of an external electric field. Here a dc field is applied to a medium through which an optical wave propagates. The change in the polarization is present in the two interacting field components effectively alters the refractive index of the medium.

2.3.6 Optical Rectification

The optical rectification is defined as the ability to induce a dc voltage between the electrodes placed on the surface of the crystal when an intense laser beam is directed into the crystal.
2.4 LINEAR AND NONLINEAR OPTICAL PHENOMENA

2.4.1 Linear optical phenomena

Nonlinear effects can also occur in gases and liquids but are mostly common in crystals. The electrons present in nonlinear crystals are bound “potential wells”, which act very much like tiny springs holding electrons in the crystal to lattice points. If an external force pulls an electron away from its equilibrium position, the spring pulls it back with a force proportional to displacement: The spring’s restoring force increases linearly with the electron’s displacement from its equilibrium position. The electric field in a light wave passing through the crystal exerts a force on the electrons that pulls them away from their equilibrium positions.

In an ordinary (i.e., linear) optical materials, the electrons oscillate about their equilibrium positions at the frequency of this electronic field. The fundamental law of physics says an oscillating charge would radiate at its frequency of oscillation, so these electrons in the crystal “generate” light at the frequency of the original light wave. Part of the energy in the light wave is converted to the motion of the electrons, and this energy is subsequently converted to light again. But the overall effect is to retard the energy as it moves through the crystal because it takes a detour into the motion of the electrons.

2.4.2 Nonlinear optical phenomena

The nonlinear materials are one in which the electrons are bound by short springs. If the light passing through the material is intense enough, its electric field could pull the electrons to reach the ends of their springs. The restoring force is no longer proportional to the displacement; it becomes nonlinear. The electrons are jerked back roughly rather than pulled back smoothly and they oscillate at frequencies other than the driving frequency of the light wave. The electrons radiate at the new frequencies, generating the new wavelengths of light. The exact values of the new wavelengths are determined by conservation of energy. The energy of the new photons generated by the nonlinear interaction must be equal to the energy of the
photons used. The energy of the two 1.064-μm photons is equal to the energy of the single 532-nm photons.

The latter half of the Twentieth Century is marked by the development of electronics based on the semiconductor industry and technology. The speed and memory of the present devices used in communication, information processing, and other areas would reach their limit there by not being able to cater the needs of the future. It is expected that the coming century would be an era for wide application of all optical technologies since, in an information society, it is necessary to rapidly transmit, receive and process a large amount of information. For large bandwidths, optical sources would serve the purpose and extensive development of optoelectronics (optics + electronics) and photonics is being undertaken. Optical devices possess the advantage and the important properties of high frequencies, broad spectral range, ultra-high speed (~few fs), the capability of parallel processing and high electromagnetic noise resistance.

Third order nonlinear optical properties provide the means to control light with light, to change the frequency of light, to amplify one source of light, switch it, or alter its transmission characteristics through a medium. The intensity dependant refractive index provides the mechanism for control in most of the third order nonlinear optical materials based devices. The discovery of the phenomena of optical bistability gives a boost to the idea of exploiting the large bandwidth (~10^{12} Hz). For the realization of photonics era, the search for novel materials is gained the most importance. The studies of optical materials with large third order nonlinearity and ultrafast response times is received increasing attention of the researchers over the last two decades for their potential applications in the area of optical communications, optical data processing, optoelectronics and many other related fields.

The progress in the realizing the potential of the materials depends on

i) Obtaining a fundamental understanding of the basic physics of nonlinear processes
ii) Based on the understanding, optimization at molecular level and later at the bulk level

iii) Using these materials to engineer viable integrated devices.

These steps include a unique blend of theoretical modeling, chemical synthesis, materials processing, complete characterization and investigation of device application [29].

2.5 NONLINEAR OPTICAL CRYSTALS

The nonlinear optical (NLO) crystals are the important material for the development of laser science and technology because there is almost a kind of materials that have functions to change frequency of laser beam and modulate it in amplitude and phase. It might be said that lasers could not be used so widely in modern science and technology today world, without nonlinear optical (NLO) crystals. Development of nonlinear optical crystals with better linear optical (LO) and nonlinear optical (NLO) properties, wider spectral transmission, and phase-matching range in particular is obviously essential for further widening the application field of lasers, the deep-UV, far IR, and even THz spectral regions. That is why many scientists working in the field today are still putting in great effort to search for new NLO crystals, even more than four decades after the invention of the laser. Advances in the development of nonlinear (NLO) optical crystals could be divided into two different areas:

- Synthesis and growth of new NLO crystals
- Improving the properties of NLO crystals

In the beginning, studies were concentrated on inorganic materials such as quartz, potassium di hydrogen phosphate (KDP), lithium niobate (LiNbO$_3$) and semiconductors such as cadmium sulfide, selenium, and tellurium. At the end of 1968, the Kurtz and Perry powder second harmonic generation (SHG) method was designed in order to find the SHG efficiency of nonlinear optical materials. In this method, a powdered sample is irradiated with a laser beam and scattered light is collected and analyzed for its
harmonic content with the use of suitable filters. For the first time, rapid, qualitative screening for second order NLO effect was possible. The stage was set for a rapid introduction of new materials, both inorganic and organic.

The second order nonlinear optical materials are used in optical switching (modulation), frequency conversion (SHG, wave mixing) and electro-optic applications, especially in electro-optic modulators. Inorganic materials are much more matured in their application to second order NLO materials than organics. Most commercial materials are inorganic especially, for high power use. However, organic materials are perceived as being structurally more diverse and therefore, are believed to have more long-term promise than in organics. The recent studies indicate the new crystals superior nonlinear optical properties and opened an area in the field of research.

For optical applications, a nonlinear material should have the following characteristics [4]:

- A wide optical transparency domain
- Large nonlinear figure of merit for frequency conversion
- High laser damage threshold
- Be readily available in large single crystals
- Wide phase matches able angle
- Ability to process into crystals, thin films, etc.
- Ease of fabrication
- Non toxicity and good environmental stability
- High mechanical strength and thermal stability
- Fast optical response time.
2.6 THE CLASSIFICATION OF NONLINEAR OPTICAL CRYSTALS

The physio-chemical properties of a material are extensively analyzed as the material is to be grown in the form of single crystal. The new materials are always investigated and the list of applications for crystals is considerably increased. After the discovery of lasers, the importances of nonlinear optical (NLO) crystals are realized. The growth of optical quality crystals is important due to the rapid requirement for the crystals. The materials required for the design of nonlinear optical (NLO) devices must fulfill certain basic conditions such as large nonlinear, appropriate transparency range, high resistance to good environmental stability. A typical second harmonic generation (SHG) active molecule must be non-centrosymmetry in nature. This symmetry requirement eliminates many materials from being SHG active and hence, at the early stage of designing and synthesizing new materials, one has to consider ways of introducing absence of centrosymmetry in the molecular structures. The classifications of nonlinear optical crystals are following:

- Inorganic crystal
- Organic crystal
- Semi organic crystal

2.6.1 Inorganic crystal

The nonlinear optical properties such as second harmonic generation (SHG), frequency up and down conversions, optical parametric amplification (OPA), optical parametric oscillation (OPO) optical emission and ferroelectric properties such as piezoelectric and pyroelectric were demonstrated in several inorganic crystals. The several inorganic crystals possess both nonlinear optical and ferroelectric properties. Lithium Niobate (LiNbO₃) is one of the most interesting inorganic materials for a wide range of applications. It possesses both nonlinear optical and ferroelectric properties. Lithium Niobate crystals are one of the most investigated materials for a wide spread and promising applications in nonlinear optical properties, parametric amplification and second-harmonic generation [30]. LiNbO₃ are widely used in optical devices such as photorefractive, holographic data storage, optical
information processing, phase conjugation and wavelength filters [31-34]. During the last decade, many new borate based inorganic NLO crystals are discovered which is greatly expanded the range of laser wavelength from the near infrared (IR) through the visible to the ultra violet (UV), deep- and vacuum-UV spectral regions. The growth of new NLO borate single crystals for UV light generation is reported by analyzed the effect of etching on the various planes for several borate crystals [35,36].

The second order nonlinear optical (NLO) properties were found in quartz crystal. Many efficient NLO inorganic crystal like KDP, Potassium pentaborate (KB5), Ammonium di hydrogen phosphate (ADP), Beta barium borate (BBO), Potassium Niobate (KNbO₃), Potassium Titanyl Phosphate (KTP), lithium tri borate (LBO) and lithium iodate (LiIO₃) were developed in the past decades for NLO applications [37,38]. Yoshida et al investigated the bulk laser induced damage threshold in KDP crystals [39]. During last few years, newly developed NLO crystals GdCa₄O(BO₃)₃ (GdCOB) and YCa₄O(BO₃)₃ (YCOB) have attracted much attention due their promising optical properties [40]. Generally the inorganic NLO crystals possess very good thermal and mechanical stability.

2.6.2 Organic crystals

The organic NLO materials play an important role in second harmonic generation, frequency mixing, electro-optic modulation, optical parametric oscillation, optical bistability, etc. [41]. Organic crystals have been extensively studied due to their nonlinear optical (NLO) coefficients being often larger than those of inorganic materials. The search for new frequency conversion materials over the past decade is concentrated primarily on organic compounds. Many new organic crystals are found, based on the predictive molecular engineering approach and are shown to have potential applications in nonlinear optics. In recent years, a considerable research is made in exploring novel organic materials for potential use in a variety of devices. The materials, which can produce green/blue laser light and could withstand high-energy light radiation, are of vital importance for their use in devices.
The organic materials are known for their applications in semiconductors superconductors and nonlinear optical (NLO) devices [42-44]. A large number of organic compounds with non-localized $\pi$ electron systems and a large dipole moment are synthesized to realize the nonlinear susceptibilities larger than the inorganic optical materials. However, their potential applications are limited by, poor chemical stability and red cut off wavelength caused by large birefringence, which results from the layer stacking of the structure and other factors [45]. The basic structure of organic nonlinear optical materials is based on $\pi$ bonding systems. Due to the overlap of $\pi$ bonding electrons, delocalization of the charge distribution takes place, which leads to a high mobility of the electron density. The extensive research shows the organic crystals exhibit nonlinear optical (NLO) efficiencies two orders of magnitude higher than those of inorganic materials [46, 47]. Hence, organic crystals are rated high as compared to inorganics in view of their large electro-optic coefficients with low-frequency dispersion, high damage resistance to optical radiation, low cost, fast and large nonlinear response over a broad frequency range, inherent synthetic flexibility and intrinsic tailor ability [48, 49]. Similarly, nowadays, Organic ferroelectrics are demanded for its diverse, useful and valuable applications such as piezoelectric devices, actuators, nonlinear optical devices, in addition to non-volatile memory, high permittivity materials and others for electronic applications. 4-dimethylamino-N-methyl-4-stilbazoliumtosylate (DAST) is one of the potential nonlinear optical crystals among the organic materials. Preparation of 4-N,N-dimethylamino-4-N-methylstilbazolium tosylate (DAST) crystal was first reported by Marder et al, in 1989 and is still being recognized as the state of the art organic nonlinear optical (NLO) crystal and nonlinear optical coefficients that are among the highest of all known materials. [50-52]. In this sequence, tartaric acid belongs to an important class of materials because of their interesting physical properties such as ferroelectricity, piezoelectricity and nonlinear optical properties [53-55].
2.6.3 Semi organic crystal

Presently, inorganic and organic materials are being replaced by semi-organics. They share the properties of both organic and inorganic materials. Recent interest is concentrated on metal complexes of organic compounds owing to their large nonlinearity [56]. Semi organic crystals are less hygroscopic compared to inorganic crystals and could be easily grown as single crystals compared to organics. Metal-organic crystals form the new class of materials under semi organics. Compared to organic molecules, metal complexes offer a large variety of structures, the possibility of high environmental stability, and a diversity of electronics properties by virtue of the coordinator mental centre [57]. Further amino acids could be used as a basis for synthesizing organic-inorganic compounds like L-arginine phosphate and its derivatives. L-arginine phosphate monohydrate (LAP) is a potential nonlinear optical (NLO) material first introduced by Chinese in 1983 [58]. LAP crystals are usually grown from aqueous solution by the temperature lowering technique. L-arginine phosphate (LAP) crystal possesses high nonlinearity, wide transmission range (220–1950 nm), high conversion efficiency (38.9%) and high damage threshold [59]. Yokotani et al. is reported the synthesis and growth of deuterated L-arginine phosphate (DLAP) crystal with a higher harmonic generation [60].

2.7 REVIEW OF Z-SCAN STUDY

The Z-scan is a study which could measure the sign and magnitude of the materials related to grown crystals. This is simple and famous method to measure third-order nonlinear optical properties. The following observations were made by various scientists in the nonlinear optical research.

Sheik-Bahae reported a simple and highly sensitive single-beam experimental technique for the determination of both sign and magnitude of nonlinear refractive index \( n_2 \) [61]. Said et al. studied the real and imaginary parts of the third order susceptibility in CdTe, GaAs, and ZnTe using Z-scan technique [62]. The Z-scan technique has been a very important technique to measure the nonlinearity because of its high sensitivity and simplicity. The Z-
scan method has been employed to measure the nonlinear absorption and refraction in a large number of direct band gap semiconductors [63-65]. Nalda de et al measured the nonlinear refractive index and absorption coefficient of the Cu:Al$_2$O$_3$ films on glass substrates by Z-scan technique [66]. Krauss et al studied the nonlinear refraction of CdS, ZnSe, and ZnS using Z-scan technique [67]. Clementi et al [68] studied the nonlinear self-focusing and absorption effects in nano structure and absorption effects. P.V. Dhanaraj reported the third order nonlinear properties of 2-Aminopyridinium trichloroacetate, (2APTC) a novel organic optical material has been synthesized and crystals were grown from aqueous solution employing the technique of controlled evaporation [69]. N.P.Rajesh reported the an organic material, nicotinium tri fluoroacetate (NTF) crystal The third order nonlinear refractive index and nonlinear absorption coefficient of (NTF) were determined by Z-scan technique [70]. P.Kalaiselvi et al reported the linear and nonlinear optical properties of L-alanine cadmium chloride (LACC) and determine the values of nonlinear optical parameter such as refractive index ($n_2$), nonlinear absorption coefficient absorption coefficient ($\beta$) and nonlinear susceptibility $\chi^{(3)}$ in LACC single crystal [71]. I.P. Bincy et al reported the negative third order nonlinear optical parameters like refractive index ($n_2$) absorption coefficient ($\beta$) and susceptibility $\chi^{(3)}$ were estimated by 2-Aminopyridinium p-Toluenesulfonate (2APPTS) organic single crystal using the Z-scan technique [72].