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Investigation on KTP performance as active element for the optical limiting and harmonic generation at IR-Vis.lasers

A Thesis
Submitted to the Deanery of College of Education at Al- Qadisiyah University in Partial Fulfillment of the certificate of M. Sc. Science in Physics

by

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﴿يَرْفَعِ اللَّهُ الَّذِينَ آمَنُوا مِنكُمْ وَالَّذِينَ أُوتُوا الْعِلْمَ

دَرَجَاتٍ وَاللَّهُ بِمَا تَعْمَلُونَ خَبِيرٌ﴾

سورة المجادلة جزء من الآية (11)

DEDICATION

To the great teacher of humanity prophet Muhammad (pbuh).

To my lady Fatima Al-Zahraa (peace be on her).

To the martyrs of Popular Mobilization.

To My family who stood by me and helped me reaching this stage.

To my brother Ali for his help.

Manar

Acknowledgment

Praise be to Allah, the Lord of all mankind and pray and peace on his propel Mohammed and his household.

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I extend my thanks and gratitude to my family, who bore the burden of study and the difficulty of continuing to complete the research

Supervisors Certification

We certify that this thesis (***Investigation on KTP Performance as active element for the optical limiting and harmonic generation at IR- Vis. lasers***) by Manar Lilo Dayekh was prepared under our supervision at the Physics Department, College of Education, and University of Al-Qadisiyah, as partial requirement for the certificate of Master in physical science.

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Certification of Linguistic Evaluation

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Abstract

The present study is related with our extensive study of the linear optical, the nonlinear optical properties, the optical power limiting effect and second harmonic generation for KTP crystal as an active element which has a dimension of 6x6x3 mm^3 by using Nd:YAG with the continuous wave (CW)at wavelength 1064 nm and double frequency 532 nmwith two powers 25 and 80 mW respectively.

The linear absorption and transmittance spectra of KTP crystal were analyzed by using an advice (UV-VIS spectrophotometer). The measurement shows that the crystal has a high transmission reached to 88% with the wavelength 1095 nm and estimated the optical constant. Also the direct forbidden optical energy gap, was found 3.8 eV.

The properties of nonlinear optics for KTP crystal had been studied by using the Z-scan technique and laser Nd: YAG of a continuous wave (CW). The nonlinear refraction index has been determined by using a close aperture of z-scan method and its magnitude was 3.59×10^{-14} cm^2/mW, 3.35×10^{-14} and 3.67×10^{-14} cm^2/mW. While the nonlinear absorption coefficient was calculated using the open aperture z-scan method were 2.71×10^{-3} cm/mW, 4.32×10^{-3} and 4.37×10^{-3} cm/mW.

The experimental results and the theoretical calculation, proved that potassium Titanyl Phosphate KTP crystal has a negative refraction index (self – defocusing), and the nonlinear absorption coefficient is established two photon absorption.

Also, the study of optical limiter for this KTP crystal by using the laser beam Z-scan technique and Nd: YAG CW double frequency 532nm for variable powers of (10-85) mW and good optical properties were obtained. The threshold powers limiting was 60mW.

The results that obtained showed that KTP crystal occurs a large nonlinear optical effects that makes it to be a good materials in the applications of the nonlinear optical devices and optical power limiting.

The ability for producing second harmonic generation of Nd: YAG CW at power of 35mW.The efficiency conversion about 13% generate green wavelength. KTP crystal is very satisfying for this type nonlinear phenomenon.

List of Abbreviations

Abbreviation	Meaning
NLO	Nonlinear Optics
SHG	Second Harmonic Generation
SFG	Sum Frequency Generation
DFG	Different Frequency Generation
OPG	Optical Parametric Generation
OPO	Optical Parametric Oscillation
QPM	Quasi-Phase Matching
THG	Third Harmonic Generation
HHG	High Harmonic Generation
XRD	X-ray Diffrectometer
UV	Ultra –Violate
KTP	Potassium Titanyl Phosphate
β-BaB2O4	β-Barium Borate
Li_3BO_5	Lithium Tribute
KTA	Potassium Titanyl Arsenate
RTA	Rubidium Titanyl Arsenate
RTP	Rubidium Titanyl Phosphate
CTA	Cesium Titanyl Arsenate
NCPM	Noncritical Phase Matching
CA	Close Aperture
TPA	Two Photon Absorption
RA, RSA	Reverse Saturable Absorption
OL	Optical Limiting
FCA	Free-Carrier Absorption
CW	Continuous Wave
LPM	Laser Power Meter

List of Symbols

Symbol	Definition	Unit
T	The Transmittance	-
A	The Absorbance	-
R	The Reflectance	-
I_R	The intensity refractive light	-
k	The Extinction Coefficient	-
Eg	The optical Energy Gap	-
n	The Refractive Index of the material	-
hυ	The Energy Photon	eV
σ	The Optical Conductivity	S^{-1}
P	The Polarization of the material	$Coul./m^2$
E	The electric field strength	V/m
$\epsilon_\circ$	The Permittivity of the Vacuum	F/m
χ	The electric Susceptibility of the medium	-
χ^1	The linear Susceptibility	-
χ^2	The Second nonlinear Susceptibility	-
χ^3	The Third nonlinear Susceptibility	-
N	The number density of dipole moments	-
ρ	The individual dipole moment	-
p	The Power of Laser beam	mw
A	The Effective Area of beam cross section	m
ω^o	The beam radius at focal point	mm
d_{eff}	The Nonlinear Coefficient	Pm/v
I	The Intensity of the laser beam at the focus	mW/cm^2
L	Crystal length	mm
p_w	Incident Power beam harmonic generation	mW/cm^2
P_{2w}	Output power Second harmonic generation	mW/cm^2
ΔK	The Phase matching	-
L_c	The coherence length	μm

c	The speed of Light	m/s
ω	The angular Frequency	rad
k	The wavenumber	m^{-1}
K_w	The wave vector of the fundamental beam	mm^{-1}
K_{2w}	The wave vector of the second harmonic beam	mm^{-1}
K_{3w}	The wave vector of the Third harmonic beam	mm^{-1}
I_ω	The Intensity of the input beam at frequency ω	mW/cm^2
$I_{2\omega}$	The Intensity of Second Harmonic Generation	mW/cm^2
$I_{3\omega}$	The Intensity of Third Harmonic Generation	mW/cm^2
q	The harmonics order	-
$z_\circ$	Rayleigh length	mm
λ	Wavelength	nm
z	The position was obtained by moving the samples along the axes of the incident beam (z-direction)with respect to the focal point	mm
T_p	Normalized crystal transmittance when mentioned at the position of maximum transmittance peak	-
T_v	Normalized crystal transmittance when mentioned at the position of minimum transmittance valley	-
ΔT_{p-v}	The Difference between the normalized peak and valley transmittance	mW
$\Delta\Phi_\circ$	The variation of phase shift	rad
L_{eff}	The effective thickness of the crystal	cm
t	The thickness of the crystal	cm
ΔT	The one peak value at the open aperture Z-scan curve	mW
β	The nonlinear Absorption Coefficient	cm/mW
α	The Linear Absorption Coefficient	cm^{-1}

h	The Plank constant	m^2kg/s
υ	The frequency	Hz
N	The number of molecules per unit area in the ground state	-
τ	The decay time	sec
σ	The cross section for the absorption	-
Nc	The intensity propagation	-
σ_c	The free carrier absorption	-
N_{eff}	The effective Nonlinear refraction Index	-
M*	The effective carrier mass	-
σ_r	The change in the index due to population of excited states	-
η_{SHG}	The Conversion efficiency of Second Harmonic Generation	-

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CHAPTER ONE

INTRODUCTION AND PREVIOUS WORKS

1.1 Introduction to Nonlinear Optics

Nonlinear optics is the study of phenomena that occurs as a consequence of the modification of the optical properties of a material system by the presence of light. Typically , only laser light is sufficiently intense to modify the optical properties of material system .The beginning of the field of nonlinear optics is often taken to be the discovery of second harmonic generation by Franken et al (1961) [1]. The theory of nonlinear optics builds on the well-understood theory of linear optics .Particularly that part is known as the interaction of light and matter. Ordinary matter consisted of a collection of positively charged cores (of atoms or molecules) and surrounding negatively charged electrons. Light interacts primarily with matter via the valence electrons in the outer shells of electron orbitals. The fundamental parameter in this light-matter interaction theory is the electronic polarization of the material induced by light [2].

The discovery of the nonlinear phenomena was the important; the second Harmonic Generation in Quartz crystal at pumping in Ruby laser beam, by Franken et al after a short period of laser discovery [3], great role in initiating a new branch of science is the nonlinear optics. This branch gets into experimental scientific field firmly, interaction of laser beam with matter, it is grew and developed hastily .Many researchers studied the effects and nonlinear phenomenon which contributed to discover many of physical phenomena [1, 2, 4]. The field of nonlinear optics became important by researchers since that time for both theoretical and practical .Then the study this .Phenomenon has been extended to include many of scientific fields like physics, chemistry, biology and engineering et al [5, 6].

The study of nonlinear optics can led to increase for knowing the basic concepts to nonlinear optical with the discovery the properties of the nonlinear different material. Thus it could be worked to improve it , developed so that it

could be suit with the practical applications in the various different technical ,like the optical communications ,the optical data store ,the optical power limiting , the optical switchers and Opt-electronic and photonic devices… etc. [4,7,8].

From the practical side, there are two types of nonlinear effects, the first relates by frequency conversion, whereas the second relates by optical modulation. Generation new frequencies from the interaction of laser beam with nonlinear matter, as the optical harmonic generation or Sum or difference - frequency -generation for incident beam on the medium and four-wave mixing are example for frequency conversion. While the electro-optic effect (Kerr effect), self-phase modulation, self-focusing and induced absorption saturation are example for optical modulation process [9, 10].

Generating new frequencies at the interaction of the electromagnetic radiation with nonlinear materials. It prescribed to Induced nonlinear polarization from the electromagnetic radiation at its interaction with the material in a suitable frequency. In the optical frequency modulation process, the laser beam work to module and develop the optical properties for material medium such as absorption coefficient and refraction index.

The typical properties for laser beam contributed more in growth and development the field of nonlinear optics. High intensity and width of narrow broading spectra with the coherence of radiation as well as to the good directional to laser beam that allows in its focusing to the smallest possible point, all make the possibility of observation the nonlinear optical phenomenon that comes out of interaction these beams with the nonlinear material more easily. It is difficult to achieve this at the interaction of the sources of normal light non coherence of low intensity with matter, The properties of materials stay, such as the absorption coefficient and refraction index, don’t effected highly in this low intensity to the incident radiations on it because there is no

effect for the atomic fields for matter. This is what happened in the processes that are known as linear optics, such as reflection, refraction and linear absorption. From practical aspect, it is possible to notice some of the effect of nonlinear optics at low intensities, for laser beam when there is increase for optical effect. If the induced dipoles vibration in the material medium at a coherent method there will be phase matching, the electromagnetic fields that portaged can be added to each other to produce a large intensity that operates on increments the nonlinear effects, if the incident light frequency on the matter suit with the effect frequency of ringing of oscillating dipoles, and multi photons processes are example for this case [11,2].

The search on nonlinear new materials are able to appear nonlinear response, improve, and develop these materials for practical uses is the most important duty in the field of nonlinear optics. This duty demands new understanding to the nature of these materials and huge study for its optical factors that has a role in the effect of its nonlinear behavior.

1.2 Nonlinear Optical Materials

It is well know from the nonlinear optics are interacted with the incident light radiation on it and generated a nonlinear response by polarization material for electric field for this radiation.

Generally, the materials are classified in to two kinds, Organic material and Inorganic material [5, 12, 13]. Organic nonlinear optic material meets large attention because of the need to obtain the necessary information to optical communication technique and photonic device. It is low cost, has optical properties, and can be adopted whereas nonlinear optical Inorganic are well know because this has mechanical stability and good chemical. In addition, they have large nonlinear optical properties such as Lithium niobate (LiNbO3),

Potassium dihydrogen Phosphate (KH2PO4), Gallium Arsenide (GaAs) and Potassium Titanyl Phosphate (KTP).

The focus in this study is on Potassium Titanyl Phosphate (KTiOPO3 or KTP) crystal, which is considered as a type of crystal that draws attention due to the nonlinear optical properties that give us good results since it represents as an active element in application of nonlinear optical properties, optical power limiter and second harmonic generation[14].

1.3 Literature Survey

Many researches and studies for Potassium Titanyl Phosphate (KTP) that includes the types $KTiOPO_4$, $KTiOPO_3$, and $KTiOPO_7$ crystal were done for what it has features and properties in the aspect of optical device and nonlinear optics. Most of the studies were on the type of $KTiOPO_4$.these studies and researches were:

John. D.Bierlein, et al (1989), [15]. The researcher has done in a study structure and crystal growth of KTP and its nonlinear and nonlinear optics, SHG, OPO, phase matching, optical electric and wave-guide. found that important material for SHG of Nd:YAG and other Nd:YAG doped laser and has been shown to have attractive properties SUM-Difference ,OPO.it also has large electro-optic cofficiencey mixing and low dielectric constant that make it potentially useful and for integrated optics application high optical damage.

P.A. Thomas, et al (1990), [16]. Notes proof the results demonstrate further proof that the short Ti-O bonds in KTP are the important structural features leading to the high optical nonlinear of KTP.

R.Desalvo, D.J.Hagan, et al (1992), [17]. Studied phase change produced by a cascaded $x^{(2)} : x^{(2)}$ process in KTP near the phase matching angle on a picosecond 1.64μm wavelength beam using the Z-scan technique .this nonlinear

refraction is observed to change sign as the crystal is rotated through the phase match angle in accordance with theory. Theory predicts the maximum small – signal effective nonlinear refractive index $n_{eff} \approx \pm 2 \times 10^{-14} \text{cm}^2/\text{W}$ for an angle detuning of ± 5 from phase match for this 1mm this crystal with measured d_{eff} of 3.1pm/v . For affixed phase mismatch, this n_2^{eff} scales linearly with length and as d_{eff}; however, for the maximum n_2^{eff} the nonlinear phase distortion becomes sublinear with irradiance for phase shifts near $\pi/4$.

D.S. Armstrong, et al (1996), [18]. Measured absolute magnitudes of the effect nonlinearity d_{eff} for seven KTP crystals. The d_{eff} derived from the parametric again of an 800nm signal wave in the sample crystals when were pumped by the frequency doubled, particularly filtered light from an injection seeded, the KTP crystal all type II phase-matched with propagation in the x-z plane, had d_{eff} values ranging from 1.97 to 3.50 pm/v. Measurements of gain as a function of phase velocity mismatch indicates that two of the KTP crystals clearly contain multiply ferroelectric domains. The uncertainly in our measurements of d_{eff} values is +5% for KTP. Through the study Q- switching results demonstrated that KTP can have widely varying values of d_{eff}.

Hiromitsu. Kiriyama et al (2000), [19]. The researcher developed an efficient four –pass quadrature frequency conversion Scheme. A high conversion efficiency in excess of 80% has been achieved for frequency doubling of 1064nm in KTP with a low fundamental laser intensity of 76μw/cm^2 a second harmonic output of 486mJwith 607mJ of the input 1064nm fundamental laser at 10 Hz.

H.P.Li, C.H.Kam, et al (2001), [20]. Investigated of third order optical nonlinearity in KTP crystal using the z- scan technique with 780nm femto second, and determined the nonlinear refraction index n_2 in KTP crystal is 1.2×10^{-15}cm^2/w it is found that the measured n_2 values in KTP crystal is constant

with the theoretical predication based on the two band models for the bound electronic career nonlinearity. From the open aperture, they have not observed no photon absorption because the band gap energy of KTP crystal that was measured (3.6) eV is greater than the energy of two 780nm photons. In addition, they found results show the nonlinear refraction KTP is dependent of the polarization of the laser beam.

Sebastin. Favre, et al (2003),[21]. Presented the results and analysis of SHG with a free-running Nd: yag slap laser and KTP crystal. The SHG efficiency is in flounced by the pulse width in this regime, a sub linear increased with laser intensity is observed at high intensity with respect to the ideal phase-matched case and the total power conversion is ultimately limited by catastrophic damage at the output fact . The second efficiency loses towards the end of the laser pulses and the sublinear behavior of the generated SHG intensity peak power of 146w at conversion efficiency of 14.6% and achieved for 15mm long KTP crystal.

E.MB, et al (2009), [22]. The studied optical transmission showed the incorporation of more defects into the KTP crystals. That grown by spontaneous nucleation that was also confirmed by micro morphological studies.

Akchav .Dhavdhry, et al (2009), [23]. Reports the measurement of refraction indices using thin plate using reflection elliposmetry a KTP crystal at different wavelength of lasers technique, and compered with different existing theoretical and experimental values of KTP and found to be in good agreement with them.

Ali. Hussain. Reshak, et al ,(2010), [24]. They have performed calculations of the linear optical properties of potassium Titanyl phosphate $KTiOPO_4$ (KTP) using the state-of-the-art FP-LAPW method. Our calculated energy gap (direct) using EV-GGA is 3.1 eV. This is in good agreement with the measured gap of

3.2 eV. We have found a good agreement between the measured and calculated reflectivity spectra and analyzed in terms of the calculated band structure, and have found that there is a considerable anisotropy between the various Components of the optical properties. KTP possesses positive Birefringence at zero energy of about 0.07.

J. Rajeev .Gandhi, et al (2011), [25]. studied characterized by XRD,UV ,and found the powder X-ray different were performed on the KTP grown to determine the lattice parameter values and also found the data of ASTM standard for $KTioPO_4$, While the UV the absorption spectra of growth crystals were studied using (Varian Carry 5E UV – visible – NIR) spectrometer in the spectral region of (200-2500) nm. The curve UV shows the cutoff wavelength at 353 nm.

J. Rajeev. Gandhi, et al(2014) [26], grown KTP crystal and studied to various characterization from through powder XRD and UV analysis, measured the nonlinear index by using a CW He-Ne laser at 632.8 nm. The measured values of nonlinear refraction of different growth planes (001), (011), (201) are in the order of 10^{-12} cm^2/W.

Arne .Potreak, et al, (2014). [27]. studied SHG of KTP and report high efficient generation of SHG radiation at a wavelength 532nm and conversion efficiency of $76\pm3\%$ was demonstrated using method to detect gray-tracking .it is revealed that the gray tracking could be a limiting factor for aspaceborne application due to the change in absorption and the resulting changes in thermal lensing .

Feng .Hui. Cao, et al (2016),[28].The researcher describes the theory of frequency doubling crystal, and gave summarization suitable for double frequency Characteristics of the nonlinear optical crystals put forward the theory of frequency doubling crystal choice Choose KTP crystal design Nd^{+3} : YAG

green laser, the stability of 532 nm laser output, the conversion efficiency of 49.3%.

Sudipta. Gangopadhyay,(2017), [29]. Analyzed of nonlinear optical properties of potassium Titanyl phosphate ($KTiOPO_4$) . The excellent properties KTP crystal has a number advantage for different applications, SHG, SFG, DFG, OPO, and an optical waveguide modulator that is nearly a factor 2 larger than that for another inorganic material.

Aims of the Study:-

The objectives of this work is to study:-

1- The properties of linear and nonlinear optics for $KTiOPO_3$ has been studied.It has different wavelengths and output powers.

2- Study the performance of $KTiOPO_3$ crystal as optical limiter with optical powers.

3- The study also tackles the possibility of generating of the second harmonic in $KTiOPO_3$ or KTP crystal.

CHAPTER TWO

THEORTICAL PART

2.1 Introduction

This chapter includes the theoretical concepts of the linear optics properties, the basic Concepts of Nonlinear Optics, the theoretical concept for KTP crystal and explains the Z-scan technique as well as the optical limiter application.

2.2 Linear Optical Properties

The study of optical properties is important in finding and limiting the absorption and transmittance of the material. In addition, it limits optical constants. Through it can be recognized on the optical energy gap and the optical conductivity [30].

1- Transmittance

Transmittance is defined as the ratio between the intensity of the transmitted beam from the matter to intensity incident beam, and can be expressed as [30]:

$$T=I_T / I_\circ \quad \ldots (2\text{-}1)$$

2- Absorption

Is the ratio between the intensity-absorbed beam with absorbed by the matter to the original intensity incident beam on it, and gives the ratio [31].

$$A = I_A / I_\circ \ldots (2\text{-}2)$$

3- Reflection

Is defined as the ratio between intensity reflective light on the matter to intensity incident beam on it, and gives in the following relationship [31]:

$$R = I_R / I_\circ \quad \ldots. (2\text{-}3)$$

The reflection joins with transmittance and absorption through the relationships [32]:

$$A + T + R = 1 \quad \ldots (2\text{-}4)$$

2.2.1 Optical constants:

1- Absorption coefficient

Absorption coefficient is the ratio of decrease in the incident beam on the matter in accordance with the distance towards spread the wave inside the matter. The absorption coefficient depends on the incident photon energy and on the properties of the matter. It could know on the nature of the electronics transition if it was direct or indirect. In addition, it could be limited by the following equation [33]:

$$I = I_{\circ} e^{-\alpha t} \quad \ldots. (2\text{-}5)$$

Where $I_{\circ}$ is the intensity incident beam, I is the transmittance of matter.α is the absorption coefficient, t is the thickness material, and after the Re-form the equation (2-5), we get [34]:

$$\alpha = 2.303A/t \quad \ldots. (2\text{-}6)$$

2- Refractive Index:

Refractive Index is defined as the ratio between the speed of the light in the vacuum to its speed in any other material medium. It also can be found through knowing extinction coefficient and magnitude of refraction value of matter.it could be calculated as the following [34]:

$$n_{\circ} = 1 + \sqrt{R}\ /1 - \sqrt{R} \ldots. (2\text{-}7)$$

Where $n_{\circ}$ is the linear refractive Index, R is the reflection value of matter

3- Extinction coefficient

Extinction coefficient is the quantity of energy that absorbed by electronics valance band at the incident of beam on the matter. i.e. (The magnitude energy that put out these electronics from the incident beams) or is the quantity of

attenuation in energy incident beams. The extinction coefficient dependents on the absorption coefficient for each matter and the magnitude of the wavelength for incident beams and counted from the ratio [35]:

$$k = \alpha\lambda/4\pi \ldots (2\text{-}8)$$

Where k is the extinction coefficient, α is the linear absorption coefficient, λ is the wavelength.

2.2.2 Optical Absorption Edge

Absorption of optical edge has three regions:

1-High absorption Region, the high absorption results from electronic transitions from the found levels of the valence band to the found levels of conduction band as in figure (2-1).Through this region knowing the forbidden energy gap and the absorption coefficient ($\alpha \geq 10^4$ cm^{-1}).As illustrated in figure (2-1A).

2-Exponential Region: The electronic transitions in this region are from the found levels in valence band to the positional levels in the valley of Conduction band or reverse of the valence band to the levels of conduction band. The absorption coefficient in this region is ($1< \alpha < 10^4$) cm^{-1} as shown in figure (1-1B). 3-Low absorption Region .The transitions in this region depends on the intensity of cases inside movement gap resulted from structural defects .The figure (1-1-C) shows the region [35].

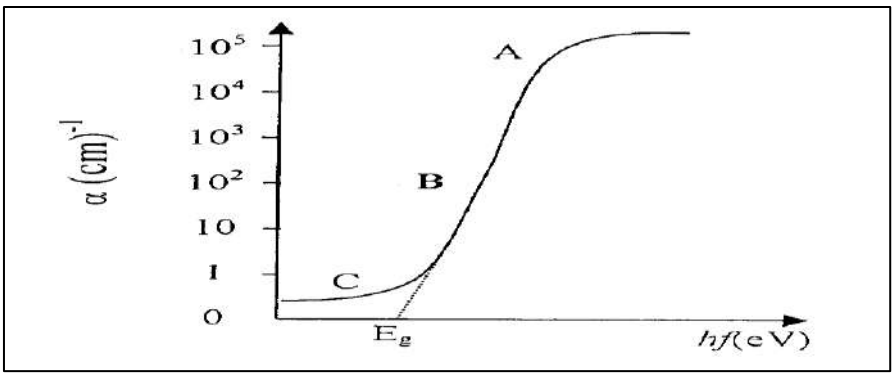


Figure (2-1) Shows the main regions of optical absorption edge [36].

2.2.3 Electronics Transitions:

Electronics Transitions are divided into direct transition and indirect transition as illustrated in figure (2-2).

2.2.3.1 Direct Optical Transition

In this transition, the valley of conduction band and top of valance band in the wave space in the same point (ΔK=0) and in this type, the absorption when it was (hυ =E_g).It happens without change in the momentum and it is two types.

Allowed direct transition: It happens when transition between highest and lowest in the valance conduct band. In this transition r = (1/2).

Forbidden Direct transition, it happens when the transition are between the neighbor points to the highest and lowest point. In this transition r = (3/2). [37].

2.2.3.2 Indirect Optical Transitions

The transition of indirect optical electronica happens when the valley of conduction band and the top of the valance band in different areas to the wave space (K) , and this type of transition happens by the phonon assistance to keep the momentum that resulted from change wave factor for the electron. These are two types of transitions:-

Allowed indirect transition: This transition is between the highest in the valance band and lowest point from the conduction band that found in different areas and r is (2) [38].

Forbidden indirect transition: This transition is between neighbor points two the highest and lowest point in the valance band and r is (3) [38], [39].

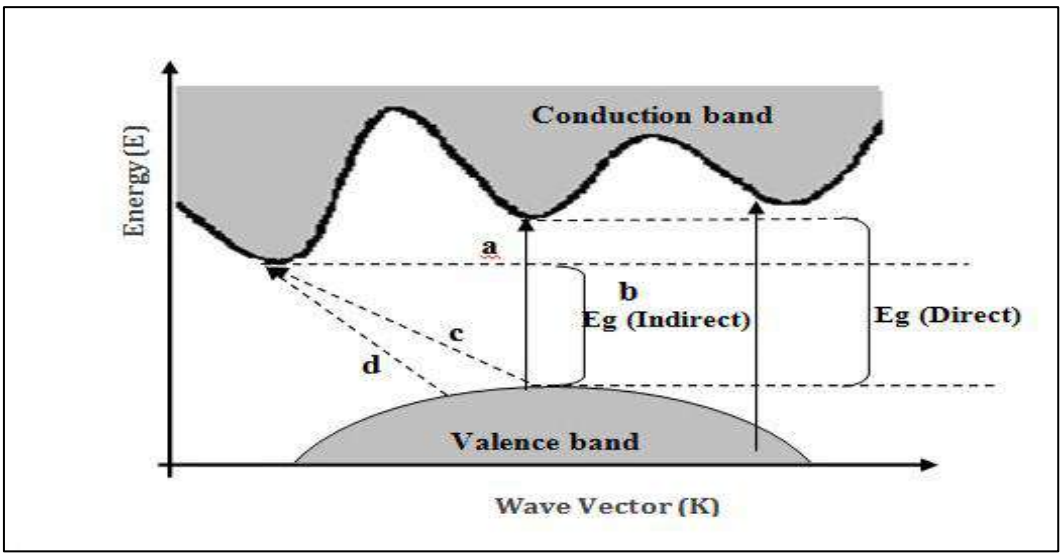


Figure (2-2) shows types of Electronic Transition [36].

2.2.4 Optical Energy Gap

Energy gap is the magnitude of power that electronics transition needs it from the top of valance band to the conduction band or the nearest position level for transition .the value of energy gap can be computed to the allowed direct transition or the forbidden and the indirect transition from (Taus equation).

$$(\alpha h\upsilon) = \beta\left(h\upsilon - E_g\right)^{r} \ldots\ldots (2\text{-}9)$$

Whereas β is the transition constant .its value one if transition is from direct or indirect [40].

2.2.5 Optical conductivity

Is the increase in the number of charges at the slip of the light beam on the material and can be calculated from the ratio as the following:

$$\sigma = \alpha\, n_{\circ} c/4\pi \quad \ldots\ldots (2\text{-}10)$$

Where σ is the optical conductivity [41].

2.3 The Basic Theoretical Concepts of Nonlinear Optics

A linear dielectric medium is characterized by a linear relation between the polarization density and the electric field, $P = \epsilon_{\circ}\chi E$, where ϵ_o is the permittivity of free space and χ is the electric susceptibility of the medium. A nonlinear relation between P and E, as illustrated in Fig (2-3), on the other hand, characterizes a nonlinear dielectric medium.

The nonlinearity may be of microscopic or macroscopic origin. The polarization density $P = \mathbf{N}\,\rho$ is a product of the individual dipole moment ρ induced by the applied electric field E and the number density of dipole moments **N** . The nonlinear behavior may reside either in ρ or in **N** [42].

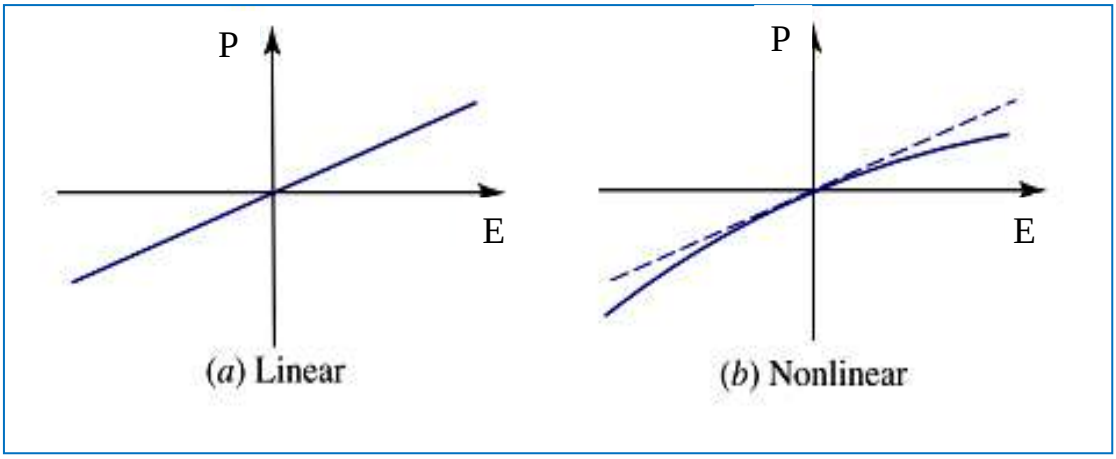


Figure (2-3) the P- E relation for

(a) a linear dielectric medium, and (b) a nonlinear medium[42].

The relation between P and E is linear when E is small, but becomes nonlinear when E acquires values comparable to interatomic electric fields, which are typically ~ 10^5- 10^8 V/m. This may be understood in terms of a simple Lorentz model in which the dipole moment is $\rho = -ex$, where x is the displacement of a mass with charge −e to which an electric force −eE is applied. If the restraining elastic force is proportional to the displacement (i.e., if Hooke's law is satisfied), the equilibrium displacement x is proportional to E. In that, case

P is proportional to E and the medium is linear. However, if the restraining force is a nonlinear function of the displacement, the equilibrium displacement x and the polarization density P are nonlinear functions of E and, consequently, the medium is nonlinear.

Since externally applied, optical electric fields are typically small in comparison with characteristic interatomic or crystalline fields, even when focused laser light is used, the nonlinearity is usually weak. The relation between P and E is then approximately linear for small E, deviating only slightly from linearity as E increases (see Fig. 2-3). Under these circumstances, the function that relates P to E can be expanded in a Taylor series about $E = 0$,

$$\mathrm{P} = \epsilon_{\circ} x^{(1)} E^{1} + \epsilon_{\circ} x^{(2)} E^{2} + \epsilon_{\circ} x^{(3)} E^{3} + \cdots \qquad (2.11)$$

Where $\chi^{(1)}$ is the linear susceptibility and $\chi^{(2)}$, $\chi^{(3)}$ . . . are the nonlinear susceptibilities of the medium. The linear susceptibility is related to the refractive index through $\chi^{(1)} = n_2 - 1$ and is responsible for the linear optical properties of the medium such as refraction, dispersion, absorption, and birefringence. The second-order susceptibility $\chi^{(2)}$ gives rise to familiar nonlinear optical processes such as second-harmonic generation (SHG), sum- and difference frequency mixing, the linear electro-optic (Pockels) effect, and, most importantly in the context of this treatment, optical parametric generation and amplification. The third order nonlinear susceptibility $\chi^{(3)}$ is responsible for the phenomena of third-harmonic generation, optical bistability, phase conjugation, and the optical Kerr effect. The susceptibilities $\chi^{(1)}$, $\chi^{(2)}$, $\chi^{(3)}$ . . . are tensors of the second, third, fourth, and higher rank, respectively [4].

In centro-symmetric media, which have inversion symmetry so that the properties of the medium are not altered by the transformation $\mathrm{r} \rightarrow -\mathrm{r}$, the P- E function must have odd symmetry, so that the reversal of E results in the reversal of P without any other change. The second-order nonlinear coefficient

d must then vanish, and the lowest order nonlinearity is of third order.

Typical values of the second-order nonlinear coefficient d for dielectric crystals, semiconductors, and organic materials used in photonics applications lie in the range $d = 10^{-24}$ -10^{-21} (C/V^2 in MKS units).

Typical values of the third-order nonlinear coefficient $\chi^{(3)}$ for glasses, crystals, semiconductors, semiconductor-doped glasses, and organic materials of interest in photonics are approximately $\chi^{(3)} = 10^{-34}$ -10^{-29} (Cm/V^3 in MKS units). Biased or asymmetric quantum wells offer large nonlinearities in the mid and far infrared [42].

2.4 Second Harmonic Generation

Franken demonstrated frequency doubling of a ruby laser using a quartz crystal in 1961[43].This was the first experiment optical frequency conversion and hence an important milestone of the investigation of the nonlinear optical properties of matter [43]. The processes of second harmonic generation involves the interaction of two waves at frequency ω to produce a wave with the frequency 2ω. It is plotted illustrated in figure (2-4) below [44].

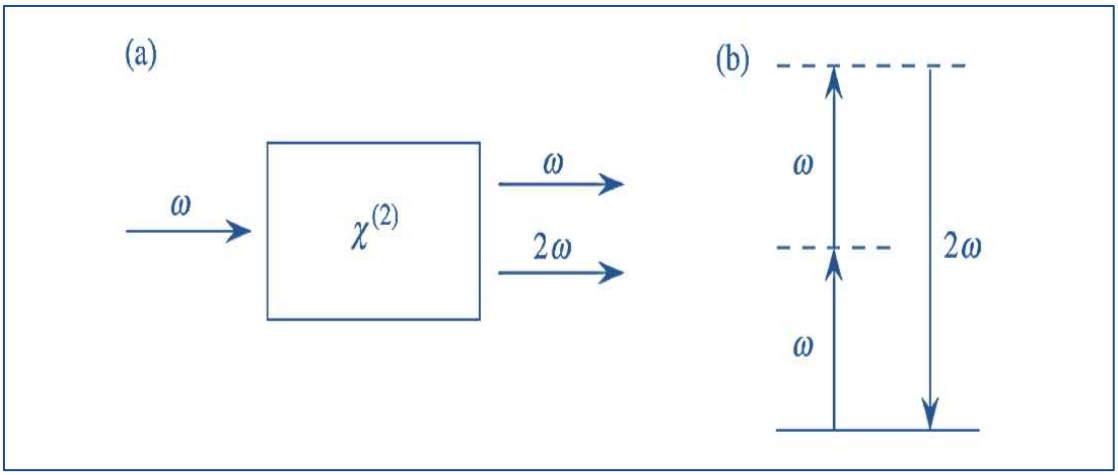


Figure (2-4) (a) Geometry of second –harmonic generation. (b) Energy –level diagram describing second –harmonic generation [1].

Figure (2-4) shows that the energies of the two absorbed photons aren't the same .This is the general case of "three wave mixing" where a third photon with the sum of the energies of the original photons are produced from the interaction of two photons combine with material . Therefore, we have two photons with the same energy level combining to produce a third with the sum of the energies of the two original photon in the case of SHG [1].

Second-order nonlinear processes can only happened in materials without inversion center. In practice, the nonlinear coefficient, d, is more frequently used than $\chi^{(2)}$ and they are interrelated by the expression:

$$d_{il} \equiv \frac{1}{2}\,\chi_{ijk} \ldots\ldots (2.12)$$

Where I, j, k are the Cartesian indices of the different frequency components polarization [45][46].

One of the most important tasks in SHG is to obtain high conversion efficiency. We know that intensity is the power (**P**) per unit Area (A) [47]:

$$I = \frac{\mathbf{P}}{A} \quad \ldots\ldots (2.13)$$

$$\mathrm{A}= \frac{\pi\omega_{\circ}^{2}}{2} \quad \ldots (2.14)$$

Where $\omega_{\circ}$ is a radius of the beam and A is effective area of the beam cross section.

The output intensity I_{SHG} has the following dependence on the effective nonlinear coefficient d_{eff} the crystal length L, the input intensity I and phase

$$I_{SHG} = d_{eff} L\, I^{2} sin^{2}\left(\frac{\Delta k L_{c}}{2}\right) \ldots\ldots (2.15)$$

When Δk the refractive indices of incident fundamental and second harmonic waves are equal and the intensity of SHG process is maximal .The characteristics dependence of SHG intensity on phase mismatch is illustrated in figure (2-5).

Therefore, the SHG efficiency can be defined as:

$$\eta = \frac{\mathbf{P}_{2\omega}}{\mathbf{P}_{\omega}} = \frac{I_{SHG}}{I_s} = d_{eff}\, LI_{\circ}\, sin^2\left(\frac{\Delta k L_c}{2}\right) \quad \ldots\ldots (2.16)$$

Where $\mathbf{P}_2\omega$ is the SH output power and $\mathbf{P}\omega$ is the incident beam power [47].

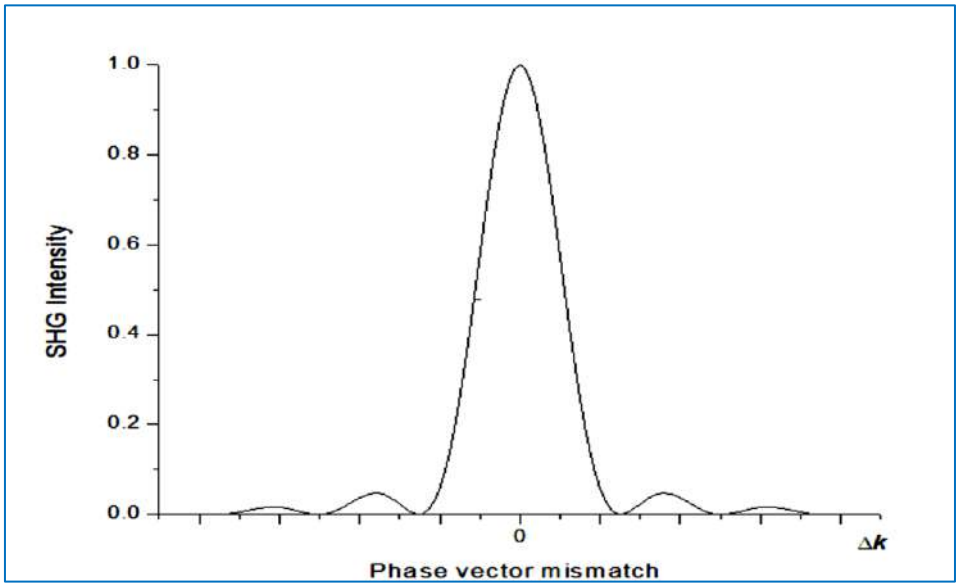


Figure (2-5) shows second harmonic output intensity vs. Δk[47].

2.4.1 Frequency Conversion

There are mainly two types of frequency conversion processes [48].The first conditions for such processes to appear is the photon energy conservation .Two waves , at frequency $\omega_1$and ω_2,incident on a nonlinear medium can combine in different ways , for SFG, the photon energies are added to create a new photon with higher energy , $\omega_3 = \omega_2 + \omega_1$. SHG is a special case of SFG when the two photons have the same frequency and come from the same laser mode, $\omega_3=2\omega_1=2\omega_2$. In the case of DFG, a photon with low energy is created by subtraction of the lower energy from the higher, $\omega_3 = \omega_1 - \omega_2$. In OPG, a pump photon is split into two lower energy photons, called the single photon of frequency ω_s and the idler photon of frequency ω_i, respectively .To keep the energy conservation, $\omega_p = \omega_s + \omega_i$. If the OPG is put into a resonator, an optical parametric oscillator (OPO) is constructed figure(2-6) illustrates these processes [46].

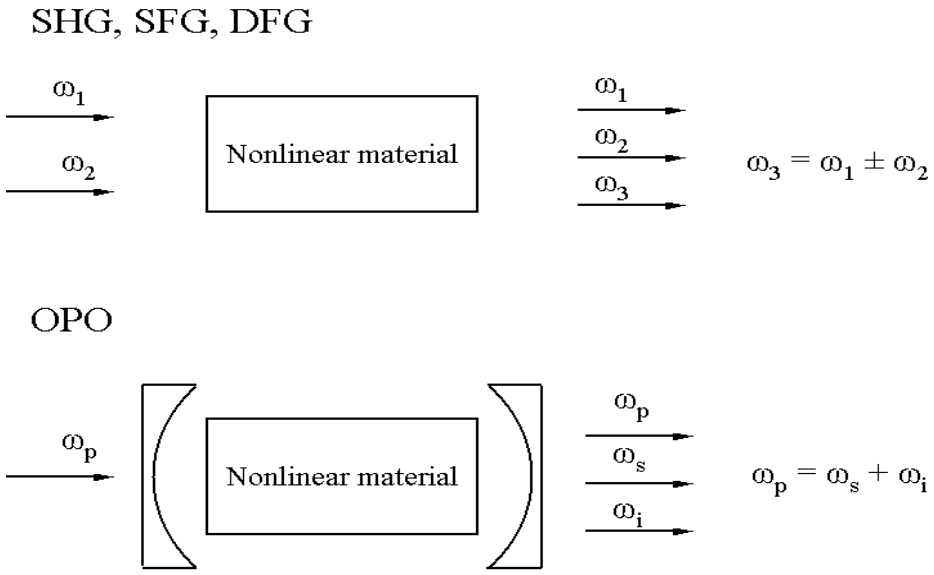


Figure (2-6) present Frequency conversion processes [46].

2.4.2 Optical Parametric Oscillator

In the process of harmonic generation, two photons combine their energies into a single photon. We have also seen that the reverse process, in which a single photon splits its energy into two photons, which will have lower energy than the incident photon, is also possible. By conservation of energy, the energy of the resulting photons must sum to the energy of the incident photon. In an optical parametric oscillator (OPO) configuration, a pump beam incident on a nonlinear crystal produces two resultant photons at different wavelengths. One wavelength, called the signal, exits the device as the output beam; the second wavelength, essentially useless, called the idler beam, stays within the cavity of the device, as depicted in Figure. The frequencies of the idler and output beam sum to the frequency of the pump beam (as required for conservation of energy). An OPO is not a laser since it does not amplify; rather, it is an oscillator only. It is, however, a coherent oscillator-producing laser light [49].

2.4.3 Phase Matching

Phase matching is a very important concept in the nonlinear optics. Phase matching technology adopted, the nonlinear effect of conversion efficiency

greatly improved [50]. It can be noted that the generated power depends on the square of the input power and also on the square of the crystal length. Dependence of the generated power puts a stringent requirement on the phase matching condition ,Δk = 0 , for optimum conversion efficiency , as illustrated in figure (2-7). The length L_c , inside the crystal which yield a phase – mismatch of π is called the coherence length .At this point , the second harmonic waves generated in the subsequent crystal segment will start to interfere will start to intensity destructively with the waves generated in the crystal [48][46].

$$L_c = \pi/\Delta k \qquad \dots (2\text{-}17)$$

For most configurations this coherence length is of the order of 1-100 μm, which severely limits the useful crystal length and, hence, the output power. One way to obtain high conversion efficiency over longer crystal lengths is to utilize anisotropic materials, birefringent properties, which yields different dispersion relations for different polarizations directions. With a suitable combination of wavelengths polarizations and propagation directions, a phase matched configuration can be obtained for the nonlinear processes .If the pair of input or output photons in the process have the same polarization the phase matching is of type –I , otherwise of the type-II[48].

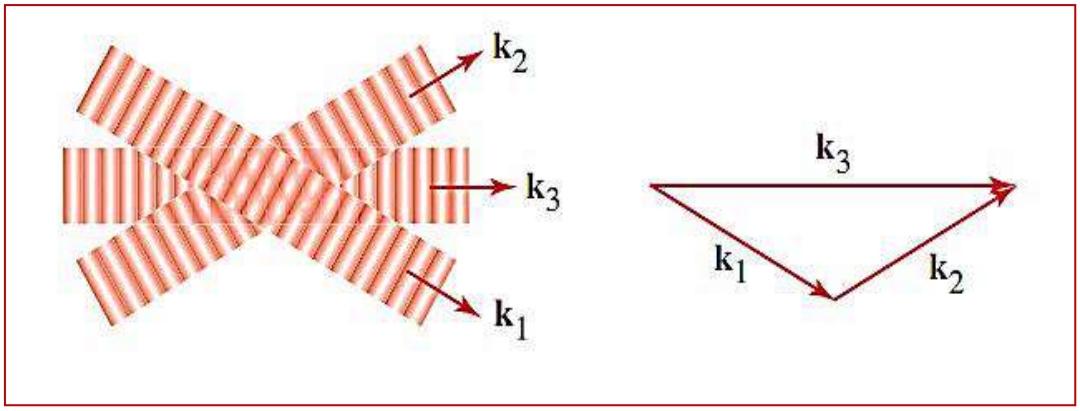


Figure (2-7) shows the condition phase matching [42]

2.4.4 Quasi-phase matching

QPM is a technique for compensation of the phase-velocity differences between the interacting waves in a nonlinear frequency conversion processes. In QPM, the accumulation of phase mismatch is prevented through a spatial modulation of the sign of the nonlinear susceptibility. Thus, the nonlinear medium is physically altered in a periodic fashion along the length of the crystal to prevent the flow of energy away from the generated wave [44]. Armstrong et al first proposed this technique in 1962 [46].

The idea is to correct the phase-mismatch between the interacting fields at regular intervals by imposing a periodic structure to the material. In second harmonic generation , If the process is not phase matched, the energy will after the first coherence length in the crystal starts to flow back from the generated field to the incident and after 2L, the generated field is back to zero energy . This energy fluctuation is repeated with a period of 2L and the generated wave will not grow with distance [46].

2.5 Third Harmonic Generation

Third order nonlinear process is applicable to either centro– symmetric or non-centro symmetric optical materials. The development of Q –switched lasers has made it possible to generate third harmonic crystals [51]. Equation $P^3 = \in_{\circ}\chi 3E^3$, represents the third order nonlinear polarization term. In this Process, an intense laser beam of angular frequency is passed through a crystal such that the beam emerging from the crystal contains the angular frequencies ω_1 of the input beam and $\omega_3 = 3\omega_1$ which is thrice the frequency of the input beam [52]. That is if infrared radiation of wavelength 1064 nm is made to incident on a crystal from an Nd: YAG laser then the output from the crystal must contain

radiation in UV region with wavelength 266nm in addition to the original 1064 nm radiation[53].

Third harmonic generation provides a mechanism for light control and hence can be used in optical switching, optical bistability and sensor protection, optical phase conjugation, four wave mixing, self -focusing , Stimulated Raman scattering, optical Kerr effect, two photon emission or absorption and coherent anti stokes Raman scattering. Third harmonic nonlinear interactions involving three frequencies degenerate input waves have been the field of active research and are used m problems of generating holographic images, which are developing rapidly and is called real-time holography [53].

Third Harmonic generation (THG) is a process of producing a wave that oscillates at the frequency 3ω by mixing three waves, each has the same carrier frequency ω as indicated in the diagram (2-8) [1]. Third harmonic generation can be performed either directly by a second harmonic process or as a two-step process involving two sequential harmonic generation step [54].

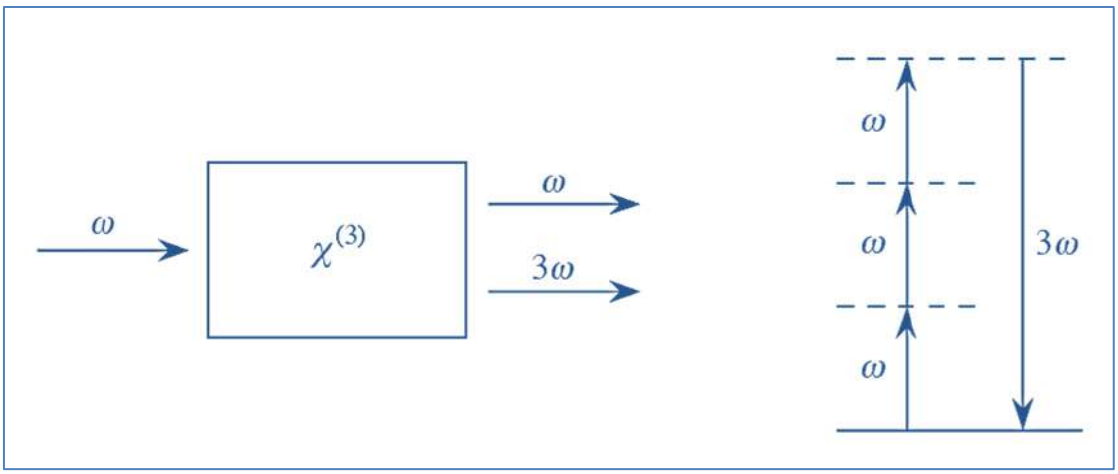


Figure (2-8) third harmonic generation: (a) Geometry of Third harmonic generation. (b) Energy –level diagram describing Third harmonic generation[1].

The absolute value of the third order nonlinear optical susceptibility $|\chi^{(3)}|$ can be expressed by the following relation [55] :-

$$|\chi^{(3)}| = \text{Re}\,(\chi^{(3)}) + \text{Im}\,(\chi^{(3)}) \quad \text{.....(2-18)}$$

Where Re ($\chi^{(3)}$): is the real part of the third order nonlinear optical susceptibility $\chi^{(3)}$, Im ($\chi^{(3)}$): is imagery part of the third nonlinear optical susceptibility $\chi^{(3)}$, were expressed through nonlinear refraction index and nonlinear absorption coefficient, by the following relation [54]:

$$\text{Re}\,[\chi^{(3)}]\,(\text{esu}) = 10^{-4}\,\frac{\in_{\circ} c^2 n_{\circ}^2 n_2}{\pi} \quad (\text{cm}^2/\text{W}) \ldots (2\text{-}19)$$

$$\text{Im}\,[\chi^{(3)}]\,(\text{esu}) = 10^{-2}\,\frac{\in_{\circ} c^2 n_{\circ}^2 \lambda\beta}{4\pi^2} \quad (\text{cm}^2/\text{W}) \ldots (2\text{-}20)$$

Where c: is the speed of the light, $n_{\circ}$: the linear refractive index, λ: is the wavelength, n_2: is the nonlinear refractive index ,β: is the nonlinear absorption coefficient.

2.6 High Harmonic Generation

The first generation of high –harmonic orders in 1987 [56] by McPherson et al who successfully generated harmonic emission up to the 17 order at 248nm in neon gas [57]. HHG provides an attractive source of coherent radiation in the extreme ultraviolet (XUV) radiation and soft X-ray region and is currently finding wide application in physics, chemistry, biochemistry and biology [57].

The HHG spectrum given a linearly polarized driving field in fig (2-9) [58].

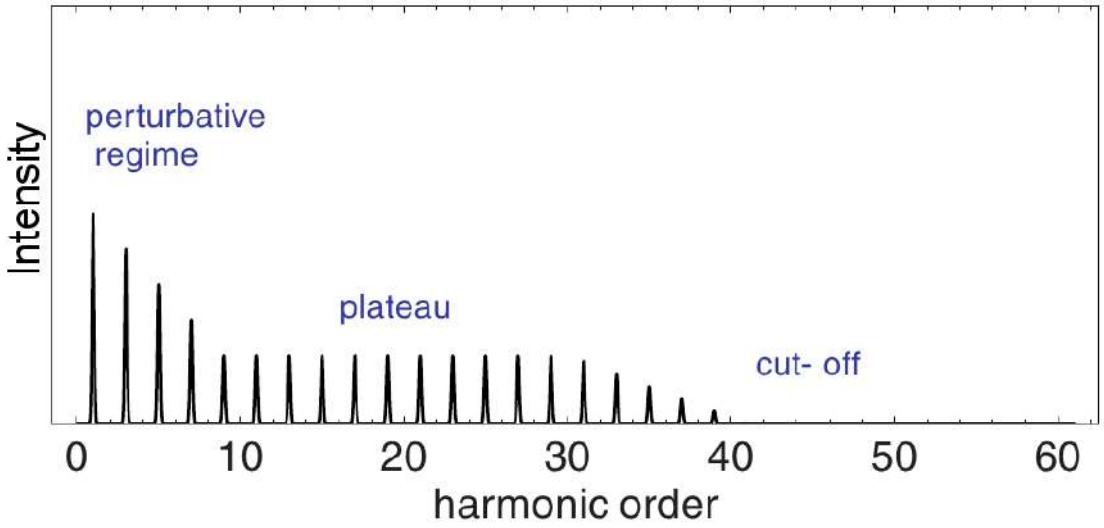


Figure (2-9) Typical HHG spectrum [57].

A typical HHG spectrum can be divided into three parts: the perturbative region at low orders, the plateau region at intermediate orders and the cutoff at region at highest orders. Perturbation theory can be used to describe the appearance of lower order harmonics (q< 9) which are produced at low intensities during the leading edge or trailing edge of the laser pulse. The harmonic yields in this region decrease as a power low [57]. High Harmonic Generation produces coherent light in high frequency regions of electromagnetic spectrum higher than any laser currently available [59].

2.7 The nonlinear refraction index and the nonlinear absorption.

The basic optical properties included in the light-mater interaction are absorption, which is defined by the absorption coefficient and refraction that is defined by the index of refraction n. When the material is irradiated, the energy of the absorbed photons makes it possible for the transition from the ground state to the excited state. This gives rise to what we call linear absorption. The further excitation may be possible due to the abundance of incoming photons; this gives rise to what we cell nonlinear absorption. The absorption of the material (α) is intensity dependent given by [60]:-

$$\alpha = \alpha_{\circ} + \beta I \quad \ldots (2\text{-}21)$$

Where, $\alpha_\circ$: linear absorption coefficient, β: the nonlinear absorption coefficient relate to the intensity.

There is also a change in the refraction index when a material is placed in a strong electric field. At high intensity, the refractive index is given by [60]:-

$$n = n_\circ + n_2 I \quad \ldots (2\text{-}22)$$

Where, n_2: the nonlinear refractive index related to the flounce, $n_\circ$: linear refractive index .The coefficients n, α are related to the intensity of the laser.

2.7.1 Saturable Absorption

Saturable absorption is the nonlinear optical process .Many material system have the property that their absorption coefficient decreases when measured using high laser intensity I of the incident laser radiation is given by the expression [1] :-

$$\alpha = \frac{\alpha_\circ}{1+I/I_S} \quad \ldots (2\text{-}23)$$

Where $\alpha_\circ$ is the initial value of absorption coefficient, and I_s is a parameter known as the saturable intensity [1].

2.7.2 Two photon Absorption

Two-photon absorption is a process of a current absorption for two photons either they have the same energy or different in energy [5, 61, 62, 63]. In this process, the two photons are absorbed by the atoms of media and moved from the ground state to the excited level passing through virtual state or virtual level. The reason is that the two photons have no enough energy to complete the transition to the excited level. As in figure (2-10).

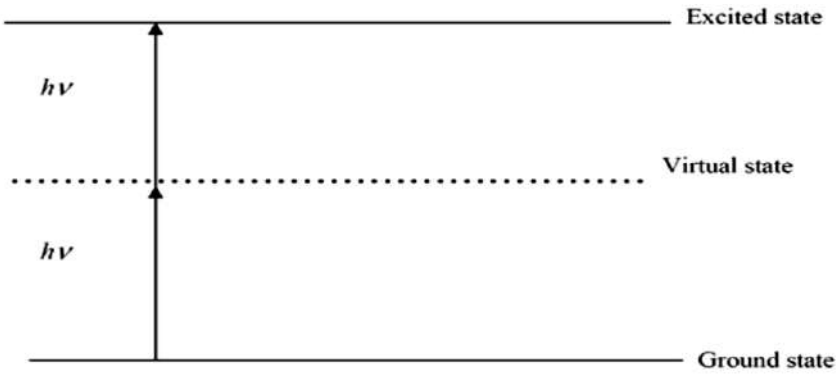


Figure (2-10) nonlinear absorption Two-photon Absorption [64].

2.8 The Self-focusing and the Self –defocusing.

When Gaussian beam passed, through a media, that has high intensity, it will make inhomogeneous change in the optical density and nonlinear refractive index to the materialistic media .When the beam progresses inside medium. Because refractive index media depends on electrical field intensity for the electromagnetic wave. Then media becomes effective on the lights that passed through the inhomogeneous optical to the range medium will lead to the change in laser beam by passing through media, either to self-narrow or to self-widening in the width of laser beam. In the first case, the intensity in laser beam maximum and the center of beam. Thus, the magnitude of refraction index larger and low down at the edges of the beam, in this case the media operates as a convex lens for beam and it is called the self-focusing. It is referred to refractive index as positive while in the second case, the magnitude of refraction index at the center beam minimum and it is increased at the edges of beam. Consequently, laser beam widening the media operates as a concave lens to beam and it is called self-defocusing and indicates refraction index as a negative [1, 2].

2.9 Z-scan Technique

There are many techniques that are used to measure the nonlinear optical properties for the mater. Z-scan technique is to be considered as the simplest method to measure the properties that described in its high sensitivity to the single light beam [65].

Sheikh Bahae et al developed a sensitive focusing measurement technique that involves focusing a laser beam through a thin sample is known Z-scan technique in 1989 [2] [4]. The Z-scan was used measure the magnitude and sign of nonlinear properties [66].It has been used to measure nonlinear optical properties of semi-conductors, dielectric, and glass, organic or carbon-based molecules and liquid crystals [67].The essential geometry is shown in figure (2-11).

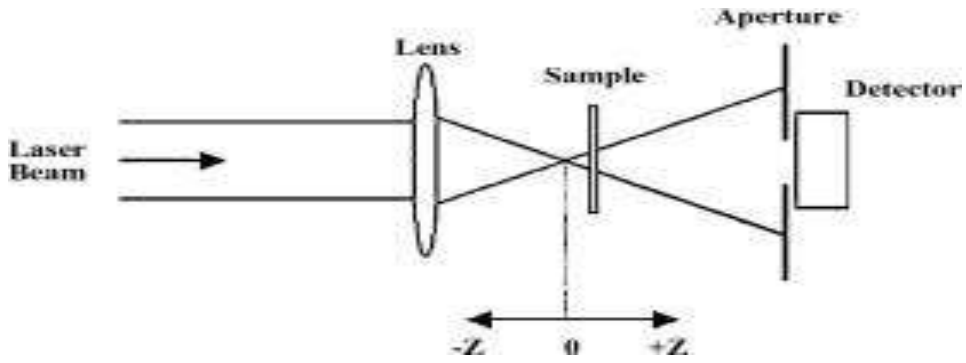


Figure (2-11) the Z-scan experimental arrangement [68]

The Z-scan works on the principle of moving the sample through the focus of the Gaussian beam in the Z-direction. The interaction of the medium with the laser light changes, as the sample moves .This because the sample experiences different intensities dependent on the sample position (z) relative the focus(z=0)[66]. The sample is usually put at the focus point of lens, and then is continuously moved along the z axis at a distance of $z_\circ$ which is expressed by the Rayleigh length $z_\circ$ [69]:

$$z_\circ = \frac{\omega_\circ^2}{\lambda}\pi \qquad \ldots. (2\text{-}24)$$

Where λ: is the wavelength, $\omega_\circ$: radius of the Laser beam.

There are two types of Z-scan: Close aperture and Open aperture. The close aperture Z-scan is used to measure the nonlinear refraction, while the open aperture Z-scan is used to measure the nonlinear absorption [69].

2.9.1 Close Aperture Z-Scan Technique

In close aperture, the sample is moved through the focal point of the focused Gaussian laser beam, the transmittance is measured through a finite aperture placed in far field, and the sign and magnitude of the nonlinear refractive index (n_2) are determined from the resulting transmittance curve [69]. To clarify the principle of the Z- scan method, we regard the propagation of focused intense laser radiation through a material under study. Regard nonlinear refraction and suppose that the medium gives a positive nonlinear addition to the refractive index (n_2). If the sample placed at a large distance from the focus (The region of negative values of z) , the radiation intensity in the medium is not sufficient to excite remarkable nonlinear refraction , and the transmission of radiation through an aperture mounted in the far-field zone stays in not variable and close to unity , as illustrated in figure (2-12). As the sample approaches the focal point, the radiation intensity increases and the self- focusing, effect appears in the medium (note solid straight lines behind the sample in figure). As a result, the far-field radiation has a larger divergence and, hence, transmission through the limiting aperture decreases [70].

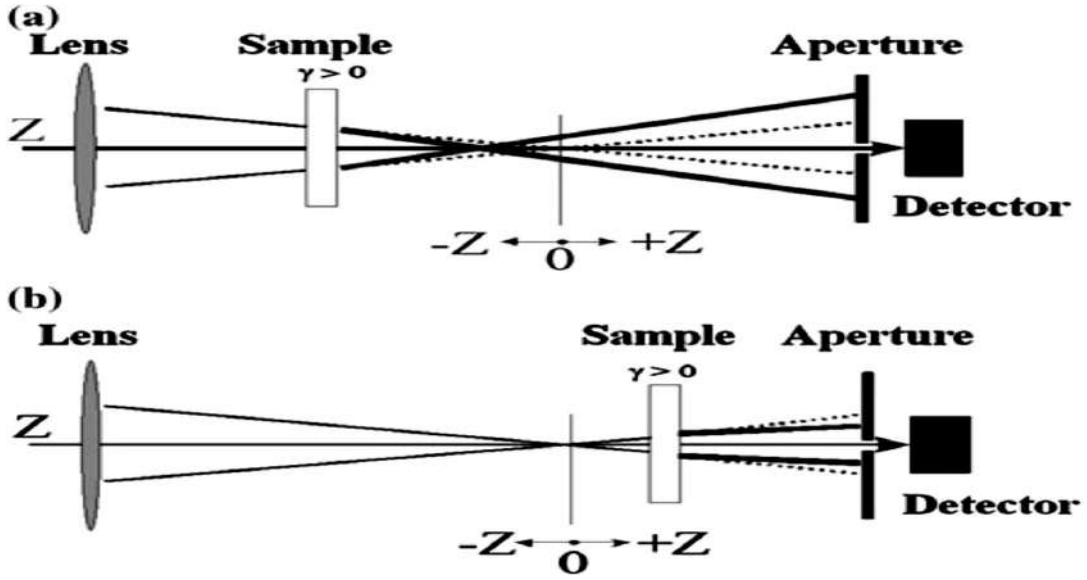


Fig (2-12) The propagation of focused intense radiation through a nonlinear self –focusing medium for a sample placed (a): in front of the focus and (b): behind it [70].

As the sample is scanned along the z-axis, transmission will decrease until the point z_1 is arrived (figure 2-12), where transmission is minimal. Decrease in the curvature of the wave front of Gaussian beam near the focus cases the general decrease in the far-field radiation divergence, and as the sample extra approaches the beam waist, transmission through the aperture increases. After the sample passes through the waist. Self-focusing reduces the far-field radiation divergence. This will go on until the influence of self-focusing on the radiation propagation through the aperture will be maximal (at the point z_2 in figure corresponding to the maximum of the solid curve). As the sample is extra displaced, the influence of this nonlinear optical process on the transmission of radiation through the aperture will decrease because of the decrease in the radiation intensity until the normalization transmission fulfill its stationary value close to unity. The medium has a positive nonlinear refractive index ($n_2 > 0$) when the maximum of the normalization of transmission follows after the minimum during Z-scan, on the contrary the medium has self-defocusing

properties ($n_2 < 0$), when the transmission maximum first appears and then minimum [69].

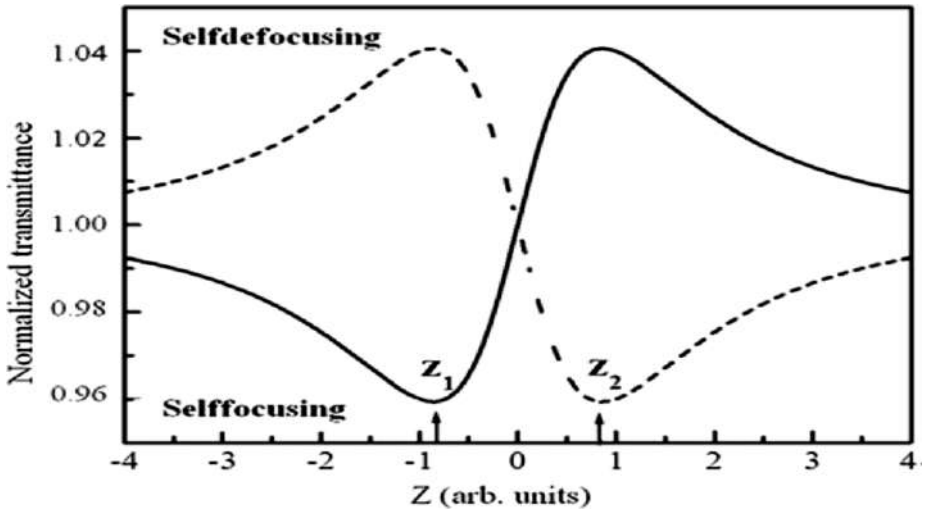
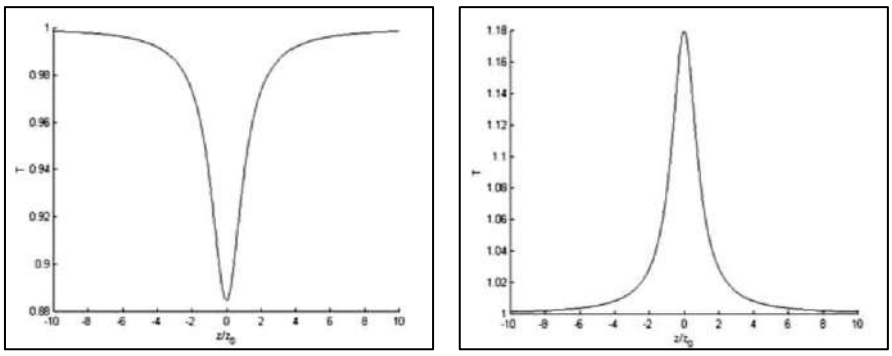


Fig (2-13) Normalized transmittance in the closed aperture graph for the media with the positive (solid curve) and negative (dashed curve) nonlinear refractive indices [70].

2.9.2 Open Aperture of Z-Scan Technique

The open aperture Z-scan method is used to measure the nonlinear absorption coefficient (β). In this case, the aperture is removed. Therefore, Z-scan technique be sensitive to the nonlinear absorption. When the absorption, increase in the sample with the increase laser beam intensity, the transmittance minimum in the focal lens as it is shown in figure (2-14a), the nonlinear absorption produces from two-photon absorption. Whereas it decrease the absorption sample with the increase of laser beam intensity ,the transmittance maximum at focal point ,as its illustrated in figure (2-14b),the nonlinear produces from saturable absorption[65]. In figure (2-14) present forms of Z-scan technique, open aperture [65].



(a) (b)

Figure (2-14) forms of Z-scan technique open aperture,

(a).Two photon absorption, (b). Saturated absorption.

2.9.3Theory of the Z-scan technique

The nonlinearity can often be evaluated from the difference between the maximum and minimum vales of the normalized transmittance; ΔT is proportion to the nonlinear phase shift $\Delta\Phi_{\circ}$,

The relation is define as [4]

$$\Delta T_{p-v} = 0.406 \left| \Delta\Phi_{\circ} \right| \qquad \ldots (2\text{-}25)$$

Where 0.406 constant quantity. And

$$\Delta\Phi_{\circ} = k\, n_2\, I_{\circ}\, L_{eff} \qquad \ldots (2\text{-}26)$$

$\Delta\Phi_{\circ}$: nonlinear phase shift, k is wave number [71], $I_{\circ}$ is the initial intensity of the laser beam at focus z=0.

$$I_{\circ} = \mathbf{P} / 2\pi\, w_{\circ}^{2} \qquad \ldots. (2\text{-}27)$$

Where $w_{\circ}^{2}$ the radius of laser beam, **P** :is the power of laser beam

Where L_{eff} is the effective thickness of the sample [72].

$$L_{eff} = (1 - \exp^{(-\alpha t)} / \alpha) \qquad \ldots. (2\text{-}28)$$

The nonlinear refraction index n_2 is given by:

$$n_2 = \Delta\Phi / k\, I_{\circ}\, L_{eff} \qquad \ldots (2\text{-}29)$$

The change of the nonlinear refraction index Δn with the laser beam intensity $I_{\circ}$ in the following relation [65]:

$$\Delta n = n_2 I_\circ \quad \text{.... (2-30)}$$

From the open aperture curve, the nonlinear absorption coefficient β can be calculate from the formula [73]:

$$\beta = \frac{2\sqrt{2}}{I_\circ L_{eff}} \Delta T \quad \text{.... (2-31)}$$

Where ΔT is the one peak or one valley at the open aperture z- scan curve.

The real part of the third order nonlinear optical susceptibility $\chi^{(3)}$, and the imagery part of the third nonlinear optical susceptibility $\chi^{(3)}$, were expressed through nonlinear refraction index and nonlinear absorption coefficient, according two relation (2-19),(2-20) respectively.

The absolute value of the third order nonlinear optical susceptibility $/\chi^{(3)}/$ can be expressed by the following relation [65] :-

$$|\chi^{(3)}| = [\mathrm{Re}\,(\chi^{(3)}) + \mathrm{Im}\,(\chi^{(3)})]^{1/2} \quad \text{... (2-32)}$$

2.10 Optical Limiting

An optical limiter is a nonlinear optical process in which the transmittance of a material decreases, when increased incident light intensity [74].The study of the optical limiting (OL) laser radiation in various materials opens the possibility of using these materials as laser shutters for protection against intense of laser radiation and is important in investigating the essential properties of nonlinear optical media [70]. One of the major potential applications of these devices is sensor and eye protection [75] .All photonic sensors, including human eye have a threshold intensity above which they can be damaged [76]. By using the suitable materials as optical limiters, allowing them to function optimally to function at higher input intensities [75]. The figure (2-15) shows the idea of ideal optical limiter has a linear transmittance at low input intensities, but above the threshold intensity its transmittance becomes constant [75].

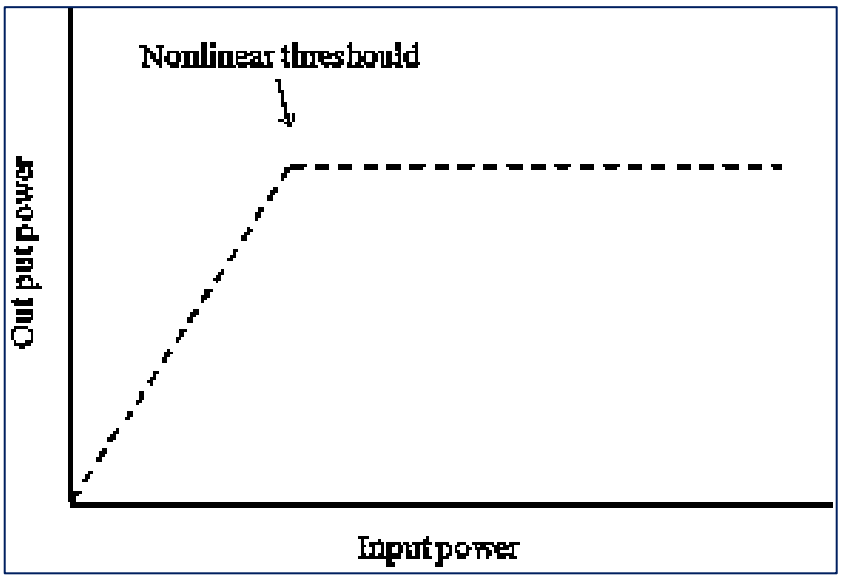


Figure (2-15) idea of ideal optical limiting [77].

2.10.1 Mechanisms Types of Optical Limiting

Optical Limiting can be achieved by means of nonlinear optical mechanisms, including self- focusing, self- defocusing, scattering, two photon absorption, free carrier, absorption, and reverse absorption in nonlinear materials [75]. The nonlinear optical mechanism can be employed for the design and performance of optical limiting devices, which can be classified into types. The first type is an energy spreading and the second is an energy absorption [74] [76]. The limiting function of an energy is based on intensity dependent change is a spatial structure of laser beam passing through a nonlinear medium [74]. This type is requirement to place an aperture in front of a detector [77]. At low input intensity levels. This change can be neglected and the whole laser beam can be detected through a property aperture in front of a detector [74]. At high intensity levels, this change becomes server that only. Small fraction of the transmitted beam can pass through the same aperture and finally detector [74]. In this type of mechanism, the nonlinear refraction that can be employed for optical limiting [77]. The optical limiting effect of refraction nonlinear optical mechanism was induced the self- focusing, self- defocusing, and scattering [75].

The second type of optical limiting is based on the intensity – dependent nonlinear attention of the laser energy in a given nonlinear materials, whereas the beam-structure changes is not so important. [74]. We only consider the second type of optical limiting power limiting device, for which the intensity dependent transmission changes in a nonlinearity absorptions material is most important [74] . In this type of mechanism, the nonlinear absorption can be employed for optical limiting was induced the two-photon absorption (TPA), reverse absorption, free carrier absorption (FCA) [75].

2.10.2The Effect of Nonlinear Absorption in OL

The energy absorption type of devices is the intensity dependent nonlinear absorption of laser energy in a nonlinear material. In this case, the beam structure change is not essentially important and therefor no aperture is needed [70]. The ideal requirements for choosing a paper nonlinearity absorbing material for OL can be described medium as follows: (1) there should be no linear absorption at the working wavelength range so that the material is highly transparent for work input light signals. (2) There is a strong dependence of the NLT on the input light intensity so that the medium is highly absorptive for intense input laser signals. (3) Threshold be a very fast temporal response of nonlinear transmission change .The following the intensity change of the input signals. (4) The optical Limiting effects should be reversible and reproducible for the input laser points [74].

Three main nonlinear absorption mechanism can be employed for OL is reverse saturable, two-photon absorption and the free carrier absorption [75, 77,78].

2.10.2.1Two – Photon Absorption (TPA)

For the two photon absorption (TPA) the interaction of electromagnetic radiation with matter [76]. Tow – photon absorption is an instantaneous nonlinearity that involves the absorption of a photon from the field to promote an electron from its initial state to a virtual intermediate state, followed by second photon, which takes the electron to its final state. Since the intermediate state is a virtual state, no energy is conserved and the system can be represented as a two or three level system.

The attenuation and excited state populations can be given by[76]:-

$$\frac{\partial I}{\partial Z} = -\alpha I - \beta I$$

and

$$\frac{\partial N}{\partial t} = -\frac{\beta I}{2h\omega} + \frac{N}{\tau}$$

$$\frac{\partial N}{\partial t} = \frac{\beta I}{2h\omega} - \frac{N}{\tau} \quad \text{..... (2-33)}$$

Here α is the linear absorption coefficient, N the number of molecules per unit area in the ground state, β is two- photon absorption coefficient and σ is a macroscopic parameter characterizing the material [76]. The cross section of the two photon absorption is determined by the two photon absorption coefficient band relies on the wavelength [75].

2.10.2.2 Reverse Saturable Absorption

Reverse saturable absorption can be noted in a system that absorbs more in the excited state than in the ground state [78].If the cross section for the absorption from the ground and the excited states are respectively $\sigma_1$and σ_2, the medium becomes more transparent when $\sigma_1 > \sigma_2$. This is because the population difference between the ground and the excited states decreases when the system absorbs light. These medium are known as reverse saturable absorbers [1]. The simplest electronic system possessing reverse saturable absorption has three

vibronically broadened electronic energy levels [78]. The RSA is a two-step and successive one photon absorption process [4].

2.10.2.3 Free – carrier Absorption

Free – carrier absorption is more prevalent in semiconductor materials, where the absorption of a photon with energy greater than the band gap energy, will promote an electron (hole) to the conduction band (valence) by absorbing additional photon. This process is often phonon assisted, although depending on the band structure and the frequency of the optical excitation, it may also be directed. The phonon-assisted phenomenon is referred to as free carrier absorption and it is analogous to ESA() in molecular system.

The free carrier absorption can readily be incorporated into the intensity propagation equation in the following form:

$$\frac{\partial It}{\partial Z} = -(\alpha + \sigma_c N_c)I \text{..... (2-34)}$$

Where α is the linear absorption coefficient, N_C is the intensity propagation and σ_c is the free carrier absorption cross section

$$\sigma_c = \frac{e^2}{n_\circ c \varepsilon_\circ m^* \omega^2 \tau} \text{ ..... (2-35)}$$

Where m* is the effective carrier mass ω is the optical frequency and τ is the free carrier decay time. It has the 1/ ω^2 dependence of high frequency conductivity and thus most important for infrared radiation in semiconductors. The free carrier density is governed by the rate equation given by [78]

$$\frac{\partial N_c}{\partial t} = \frac{\alpha_\circ}{h\,\omega} I - \frac{N_c}{\tau} \text{ ..... (2-36)}$$

2.10.3 The Effect of Nonlinear Refraction in OL

The energy –spreading is the intensity dependent change in the beam structure and the spatial energy distribution of a laser beam passing through the nonlinear medium. At low intensity level, this change can be neglected and all laser beams can be detected through a properly placed aperture in front of a detector. Whereas at high intensity levels, this change becomes more severe and only a small portion of beam energy can pass through the aperture and be finally detected [74, 77]. From Kramers – Kronig relations, we know that all materials exhibiting nonlinear refraction. This can usually be expressed as:

$$n_{eff} = n_{\circ} + n_2 I + \sigma_r N_{ex} \quad \ldots\ldots (2\text{-}37)$$

Where n_2: describes instantaneous index changes properties to incident irradiance and σ_r : describes the change in the index due to population of excited states. Third –order optical nonlinearities, n_2 is required to the TPA coefficient, β, by Kramers –Kronig relations. As a focused beam has a spatially varying irradiance, then the induced index change varies across the beam profile, causing the beam to be strongly distorted upon propagation. Near focus, the beam is usually brightest in the center, so for a negative index change (where the index decreases with increasing irradiance or flounce), the nonlinear material will behave like a lens with negative focal length, and the beam is defocused. This process is referred to as self-defocusing. If the sign of the index change is positive, self-focusing results [4].

2.10.4 Nonlinear Scattering

The presence of irregularities in the medium that can be associated with variation of the refractive index, results in the scattering of light, i.e. give rise to the extinction of the light beam. When the power of the incident light is high

enough to produce or modify scattering cross section increases with the intensity suppressing the intensity of the transmitted light leading to OL [76].

In the case of nonlinear scattering, the formation of small scattering centers in the device can be thermally induced local changes (such as density or in the form of phase changes) or, by utilizing a heterogeneous material whose components have matching refractive indices for low input intensities, while for higher power the indices become mismatched, and therefore nonlinear scattering occurs. Increased scattering at higher intensity limits the directly transmitted intensity [75].

2.10.5 Comparison Ideal Optical Limiter and Realistic Optical Limiter

The ideal optical limiter is illustrated in figure (2-16 (a)).For some materials, e.g., two photon absorbers, this transmittance may be near 100 %, and the input –output curve would have a slope of 45^{o}. On the other hand, RSA materials require a certain amount of linear absorption, and thus the input- output slope in the linear regime$< 45^{o}$. At some critical intensity or threshold in an ideal limiter, the transmittance changes abruptly and exhibits an inverse intensity or fluent dependence.

In a real material, illustrated in figure (2-16 (b)), this transition is not so abrupt. The definition of threshold is not precise. In addition, in a real material, the output is not always clamped at a constant value, but the input-output slope is decreased as shown in figure (2-16(b)). Thus, if the input-output slope is nonzero, at a some input above threshold the device will faill to provide protection. In some cases, the material itself may be damaged if its damage threshold is below this point, or the intensity/ flounce dependent transmittance may approach a constant asymptote so that the input-output slope again increases[2][77].

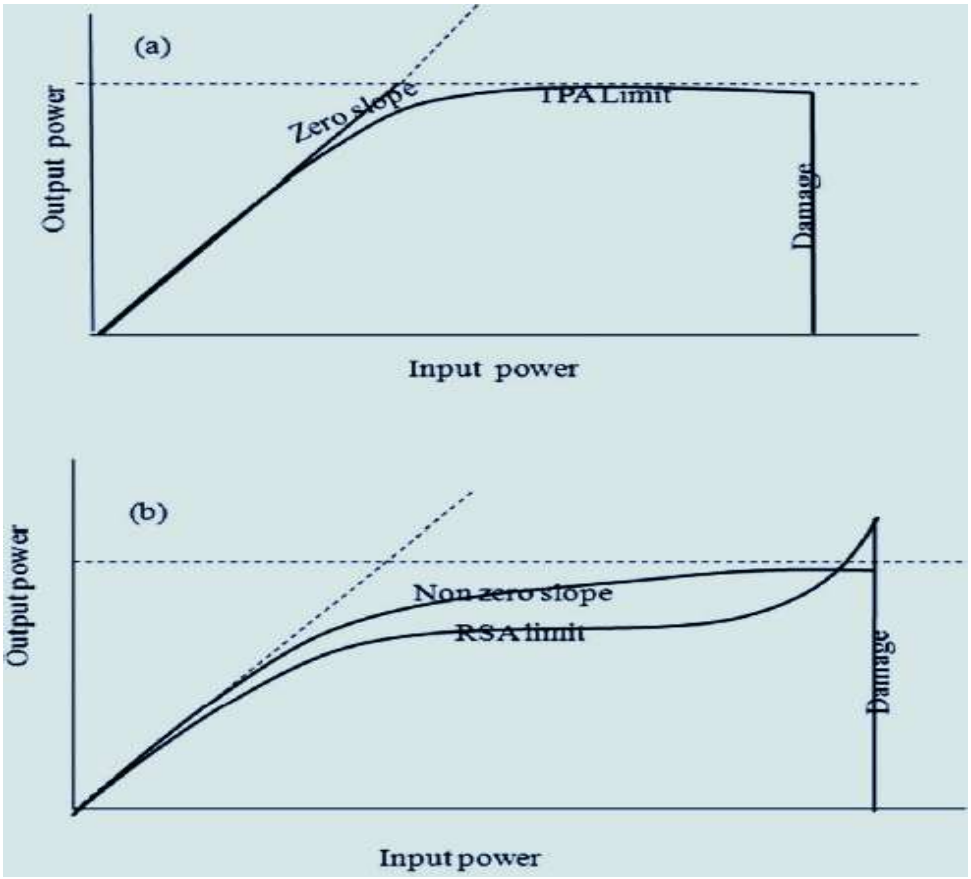


Figure (2-16) Characteristics of an optical limiter (a) Ideal optical limiter (b) realistic optical limiter, repotted form [2][77].

2.11 Main Characteristics of Potassium Titanyl phosphate crystal.

KTP was first developed in the end of the 1970[79]. It got a new attention. Tordjman et al analyzed the crystal structure in details in 1974 ,and in 1976 researcher at Dupont Inc ,USA, started to investigate KTP's nonlinear optical as well as its mechanical properties[79].The KTP is biaxial crystal[80]. The crystal possesses high nonlinear, it has a high resistant to optical [79], and has been widely used in a various nonlinear-optical applications in particular in SHG and OPO devices [79]. KTP crystal is orthorhombic and belongs to the acentric point group mm2, with only slightly different lattice parameters [81]. For this point group symmetry class, there are five non-zero nonlinear coefficients, d15 = 6.1pm/V, d31=6.5pm/V, d24=7.6pm/V, d32=5.0pm/V, and d33= 13.7pm/V [81] [82] .These values are still considerably higher than many other nonlinear

materials including urea, β-BaB_2O_4 and LiB_3O_5 [82]. There are two main techniques presently used to grow a KTP crystal, which includes the hydrothermal and flux method [25]. Potassium Titanyl Phosphate (KTP) crystal is nonlinear crystal .The famous type is $KTiOPO_4$.Another type such as $KTiOPO_7$, $KTiOPO_3$.All these types are Orthorhombic and belonged to it with only slightly different lattice parameters.

2.11.1 Growth Crystal

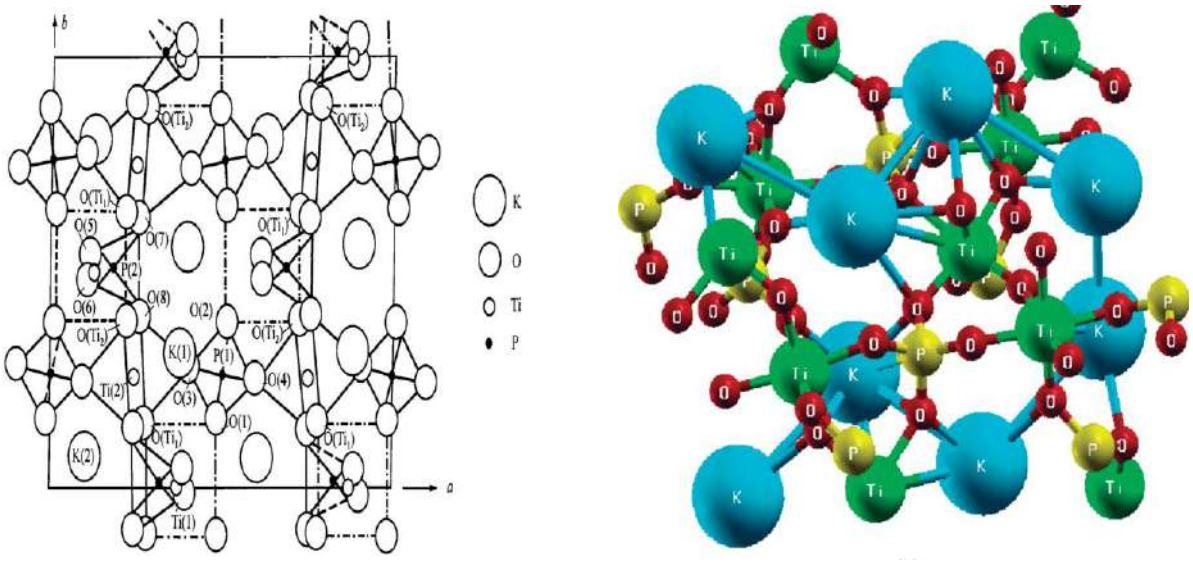
There are mainly two methods for growing KTP. In the first , the crystals are growing from a solvent that contains the dissolved components necessary to form the crystal .The solvent is called flux and has given name to the processes ,flux growth. If the solvent only contains the material which constitutes the crystal is named a self-flux .The self-flux is kept in Pt-containers at a high temperatures (800-950C) and the growth started in the supersaturated melt and continuous during cooling. The stability of the temperature is an important issue for this processes .It should be better than +/-0.05 Kat the crystal position otherwise flux inclusions and growth striations might appear .Cheng at el, have synthesized KTP,KTA,RTA,RTP and CTA crystals using the method of self – flux growth[79].

The other method is called hydrothermal growth. Here the synthesis takes place in a container called autoclave at high temperatures and pressures, where aqueous solvents are used to dissolve and recrystallize the material .Typical parameters foe hydrothermal temperatures of 400-500C and pressures of 1-2kbar.Growing KTP by a self-flux process is faster than using the hydrothermal technique[79].

2.11.2 Structure $KTiOPO_4$ Crystal

KTP is orthorhombic and belong to the centric mm cemetery group. It is also a member of the space group Pna21, which implies that the

Crystallographic direction a, b, c, corresponded to the optic axis x, y, z with c being the polar axis. KTP's structure consists of distorted TiO_6 octahedra , which is linked at two corners to form chains . Ti atom is displaced from the center of the octahedron, leading to one long , one short and for medium Ti-O bonds in the structure .These alternating lengths of the Ti-O bonds case a net c-directed polarization in KTP .The TiO_6 chains are separated by slightly distorted PO_4 tetrahedra , see figure (2-17). The K^+ ions are weakly bonded to both the tetrahedra and octahedra and can a copy to different sites – other an eight –fold or nine-fold coordinated. Further, the actions can be diffusion mechanism travel through channels, which exist along the c direction .The diffuse by a vacancy hopping mechanism that cases the ionic conductivity along the c axis to be several orders of magnitude higher than in the other direction [46].



(a) **(b)**

Figure (2-17) the KTP crystal structure [46] [24].

2.11.3 Potassium Titanyl Phosphate $KTiOPO_3$ (KTP) crystal

$KTiOPO_3$ single crystal is orthorhombic and belongs to the centric point group mm. the space group of KTP is Pna21 and the lattice parameters are a = 6.404 A°, b = 10.616 A°, c = 12.814 A°. KTP is optically biaxial crystal; the refractive indices for the principle axes of the material are all different, $n_x \neq n_y \neq n_z$ [79]. It has been widely used frequency doubling of the near –IR from Nd-YAG lasers into the green and for near –IR OPOs. The figure (2-18) shows the transmission for the potassium [83]. KTiOPO3 has wide range of transparency between (350-45000) nm [84].

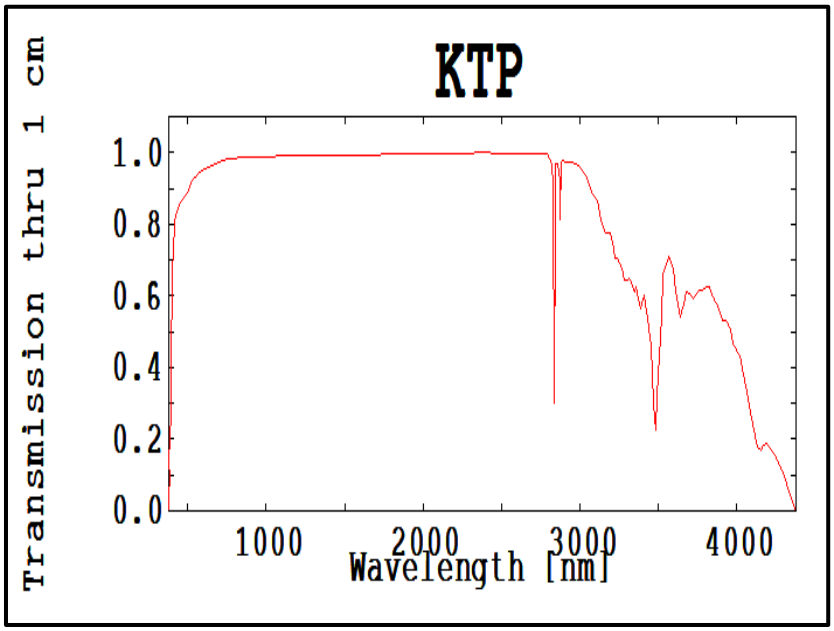


Figure (2-18) Transmission for KTP crystal [77].

The magnitudes of the d-coefficients to be d15 = 6.1pm/V, d31=6.5pm/V, d24=7.6pm/V, d32=5.0pm/V, and d33= 13.7pm/V. The expressions for d_{eff} for type II interaction to

$$\left| d_{eff} \right| = (d24 - d15)\sin 2\theta \sin 2\phi - (d15\sin 2\phi + d24\cos 2\phi)\sin \theta \quad \ldots(2\text{-}38)$$

The angle θ is measured relative to the z-axis and ϕ is measured in the xy-plane from the x-axis[81].

2.11.4 Application for SHG and SFG of Nd : Lasers

KTP is the most commonly used material for frequency doubling of Nd : YAG and other Nd –doped lasers , particularly when the power density is at a low or medium level. To date, extra –and intra –cavity frequency doubled Nd:lasers using KTP have become a preferred pumping source for visible dye lasers and tunable Ti: Saphire lasers as well as their amplifiers .They also useful green sources for many research and industry applications[84] .

More than 80% conversion efficiency and 700 mJ green laser were obtained with a 900 mJ injection –seeded Q-switch Nd: YAG Laser by using extra –cavity KTP .

8W green laser was generated from a 15w LD pumped Nd:YV04 with intra –cavity KTP.

200 mW green outputs are generated from 1 W LD pumped Nd : YV04 lasers by using THATSHIGH's 2x2x5 mm KTP and 3x3x1 mm Nd : YV04.

2-5mw green outputs are generated from 180 mw LD pumped Nd : YV 04 and KTP glued crystals . For more details, please refer to Glued Crystals. KTP is also being used for intra cavity mixing of 0.81μm diode and 1.064μm Nd: YAG laser to generate blue light and intra cavity SHG of Nd: YAG or Nd: YAP lasers at 1.3μm to produce red light.

2.11.5 Application for OPG, OPA and OPO

As an efficient OPO crystal pumped by a Nd : laser and its second harmonics , KTP plays an important role for parametric sources for tunable outputs from visible (600nm) to mid-IR (4500nm) as shown in fig (2- 19) .

Generally, KTP's OPOs provide stable and continuous pulse outputs (signal and idler) in Fs. with 108 Hz repetition rate and a mW average power level. A KTP's

OPO than are pumped by a 1064 nm Nd: YAG laser has generated as high as above 66 % efficiency for degenerately converting to 2120 nm[84].

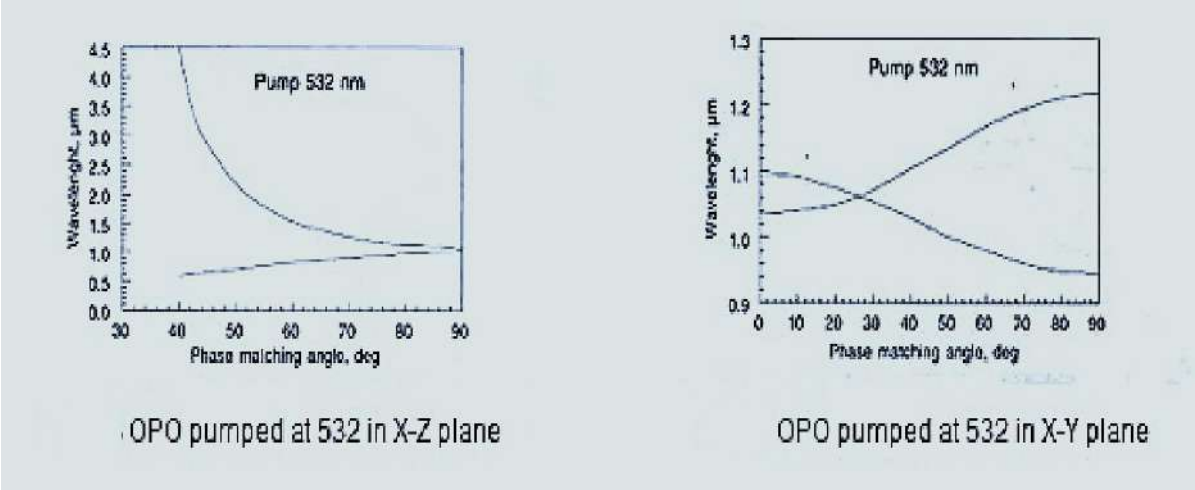


Figure (2-19) pumped at 532nm[84].

The novel developed application is the non- critical phase –matched (NCPM) M KTP OPO / OPA pumped by the X-cut KTP crystal. As shown in Fig 5 for pumping wavelength range from 0.7 µm to 1.45µm, the outputs can cover from 1.04 µm (signal) and from 2.15µm to 3.2µm (idler). More than 45% conversion efficiency was obtained with narrow output bandwidth and good beam quality[83].

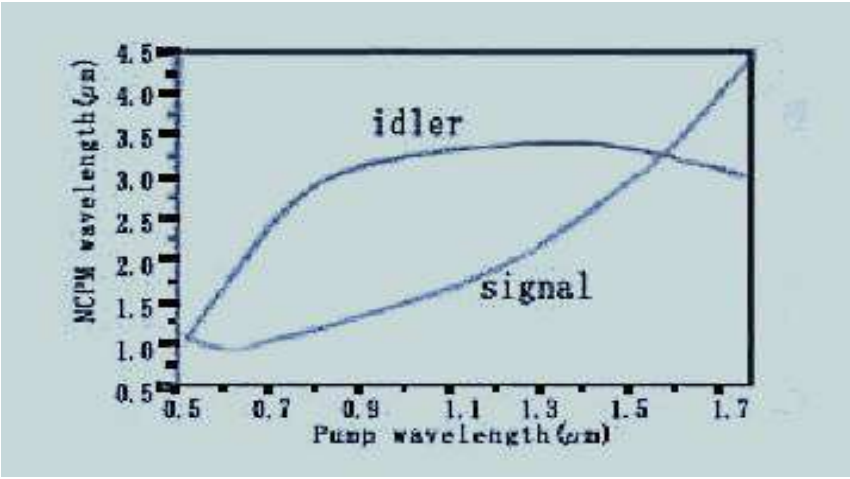


Figure (2-20) Type II KTP crystal NCPM OPO[84].

CHAPTER THREE

EXPERIMENTAL PART

3.1 Introduction.

This chapter focuses on the materials , devise that are taken to be involved for the current work and it demonstrates the Z-scan technique to determine the nonlinear optical properties and optical limiter .As well as explains the second harmonic generation .the measurement were achieved at wavelength (1064nm ,double frequncy532nm).

3.2 Spectrophotometer

To determine the optical properties for KTP crystal, The Mega -2100 Double Beam UV-Vis-IR Spectrophotometer) has been used (Mega (UV-2100) at wavelength range from 200 to 1100 nm. The transmission and absorption spectra measured as function for wavelength. From absorption spectra, the calculation of absorption coefficient and calculation of optical constants that represented by equation (2-6) refractive index, by depending the equations (2-7) respect that special for each constant.

Figure (3-1) UV-Visible Spectrophotometer

3.3 Nd: YAG Laser

In this work, the CW Nd: YAG laser was used. It composes of wavelength 1064nm at power (35mw) and double frequency 532nm at powers (85,25mw). The characteristics of Nd: YAG laser listed the below table (3-1).

Table (3-1) Characterization of Nd: YAG laser.

Physical and Chemical Properties	**Optical properties**
Chemical Formula Nd:$Y_3A1_5O_{12}$	Lasing Wavelength (532,1064) nm Radiative Lifetime 550 ms
Melting Point 1970°C (2243K)	Divergence angle 0.625 mard
Density 4.56 g/cm_3	Linewidth 0.6 nm
Refractive Index 1.82	

3.4 Detector

Laser power meter Lp1, detector LpoA1 and Laser power meter were used. Laser power meter LPO1 that has 40µw -40mw .OPT-1A power in dicator scale from 20mw – 200mw, while laser power meter has its scale from 100nm - 1100nm.

3.5 The Potassium Titanyl Phosphate Crystal

KTP Potassium Titanyl Phosphate ($KTiOPO_3$ or KTP) Crystals are widely used in both commercial and military laser including laboratory and medical systems, range finders, lidar, optical communication and industrial systems.

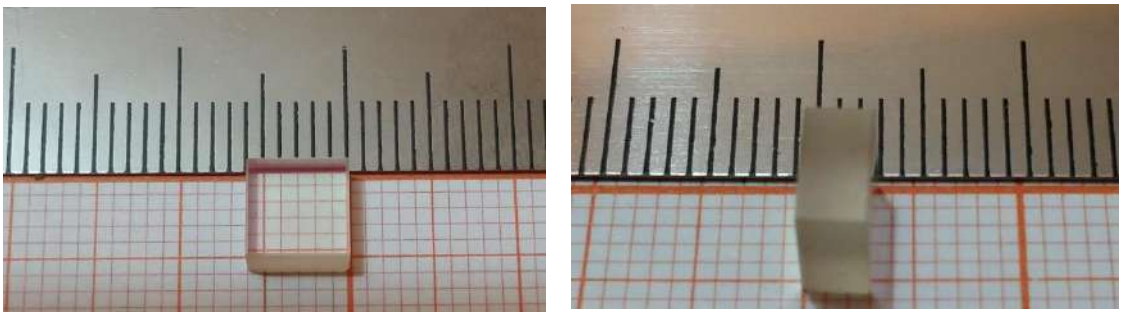


Figure (3-2) The ($KTiOPO_3$) crystal 6*6*3mm^3

Table (3-2) shows Specifications for KTP Crystal

Dimension Tolerance	$(W\pm\mathbf{0.1}\boldsymbol{mm}) \times (\boldsymbol{H}\pm\mathbf{0.1}) \times (\boldsymbol{L}+\mathbf{0.5}/-\mathbf{0.1}\boldsymbol{mm})(\boldsymbol{L}\geq \mathbf{2.5}\boldsymbol{mm})(\boldsymbol{W}\pm\mathbf{0.1}\boldsymbol{mm}) \times (\boldsymbol{H}\pm\mathbf{0.1}\boldsymbol{mm}) \times (\boldsymbol{L}+\mathbf{0.1}/ -\mathbf{0.1}\boldsymbol{mm})(\boldsymbol{L}<\mathbf{2.5}\boldsymbol{mm})$
Angle Tolerance	$\Delta\boldsymbol{\theta}\leq\mathbf{0.25}°\ \Delta\emptyset\leq\mathbf{0.25}°$
Clear Aperture	90% of central area
Chamfer Chip	≤0.2mm@45 ≤ 0.1mm °
Damage Threshold[GW/cm^2]	> 0.5 for 1064nm,TEM00,10ns,10Hz(AR-coated) > 0.3 for 532nm,TEM00,10ns,10Hz (AR-coated)
Flatness	< λ/8@ 633nm
Parallelism	<20"
Perpendicularity	≤ 5,
Surface Quality [S/D]	< 10/5
Wave front Distortion	< λ/8@ 633nm
Coating	AR coating available upon request No visible scattering paths or centers [inspected by 50mw green laser]
Interior Quality	

3.6 Z- Scan Technique Experimental

The nonlinear optical properties for Potassium Titanyl Phosphate ($KTiOPO_3$ or KTP) crystal is explained through z-scan measurements to determine the nonlinear refraction index and the nonlinear absorption coefficient. The z- scan experiment illustrates in the figure (3-3).

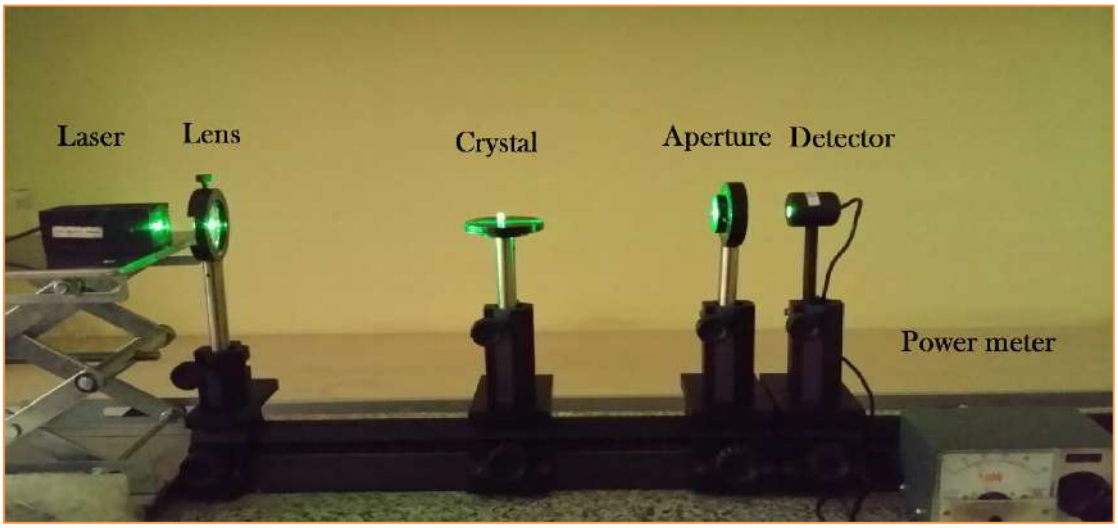


Figure (3-3) the Z-scan technique set –up experimental in 532nm

In figure (3-3) shows the elements z- scan experiment. In this experiment consisted of the laser frequency doubler Nd: YAG 532 nm by a power (25) mw and the lens which for focal length 15 cm. The crystal with dimensions ($6x6x3mm^3$) was scanned using transition system along direction z- axes through the focusing area. As well as the aperture place a front detector which its diameter 1mm. The detector was placed at the far field of laser beam.

In figure (3-4) shows the elements z-scan experiment consisted of the Nd: YAG with wavelength 1064 nm at a power 35 mw. Then we used the laser He / Ne which alignment in perpendicular to the set-up .After that mirror reflected the laser beam inside the set-up. He /Ne laser reflected and transmits laser beam toward Nd: YAG laser source. We used the lens with focal length 15 cm too, and the aperture placed between the crystal and the detector, which measures the transmission. The measurements of the normalization transmissions varies

crystal position, allow the determination of the nonlinear refraction index and the nonlinear absorption coefficient.

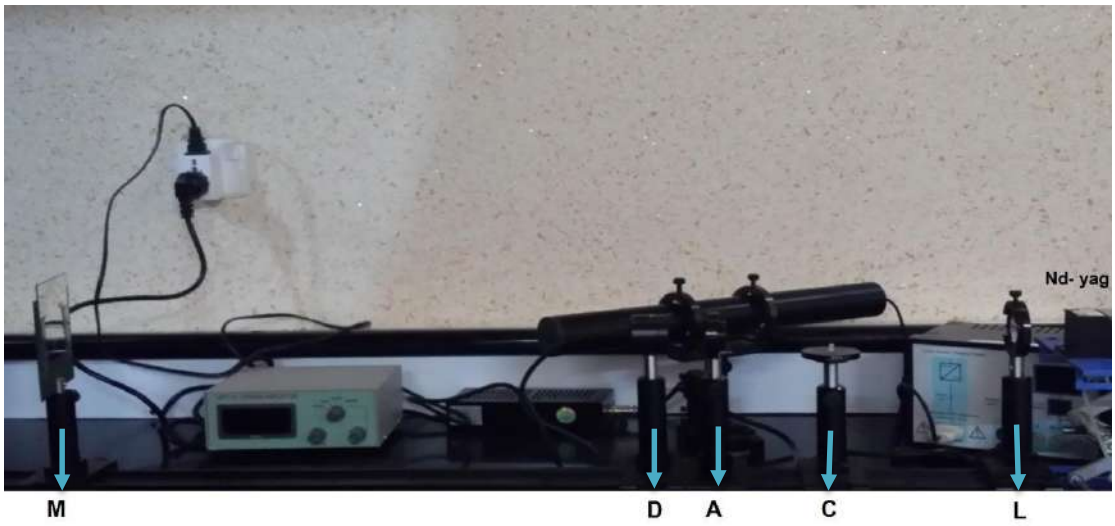


Figure (3-4) picture of the Z-scan set –up experimental

L: Lens, C: Crystal, A: aperture, D: detector, M: mirror

3.7 Experimental Set – up of the Optical Limiting

The optical Limiting experiment was performed for $KTiOPO_3$ crystal using continuous wave second harmonic Nd: YAG laser of wavelength – 532nm and power 85mw. The experimental limiting set-up for the demonstration of Optical Limiting behavior as shown in Fig (3-5, 3-6).

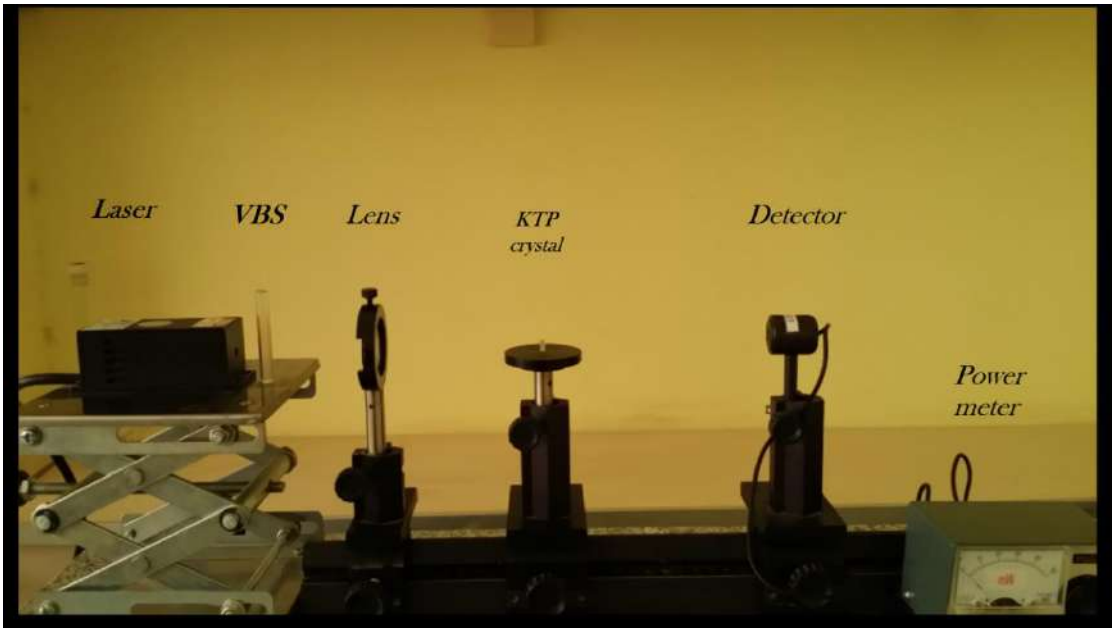


Figure (3-5) set –up of Optical Limiting for ($KTiOPO_3$) crystal

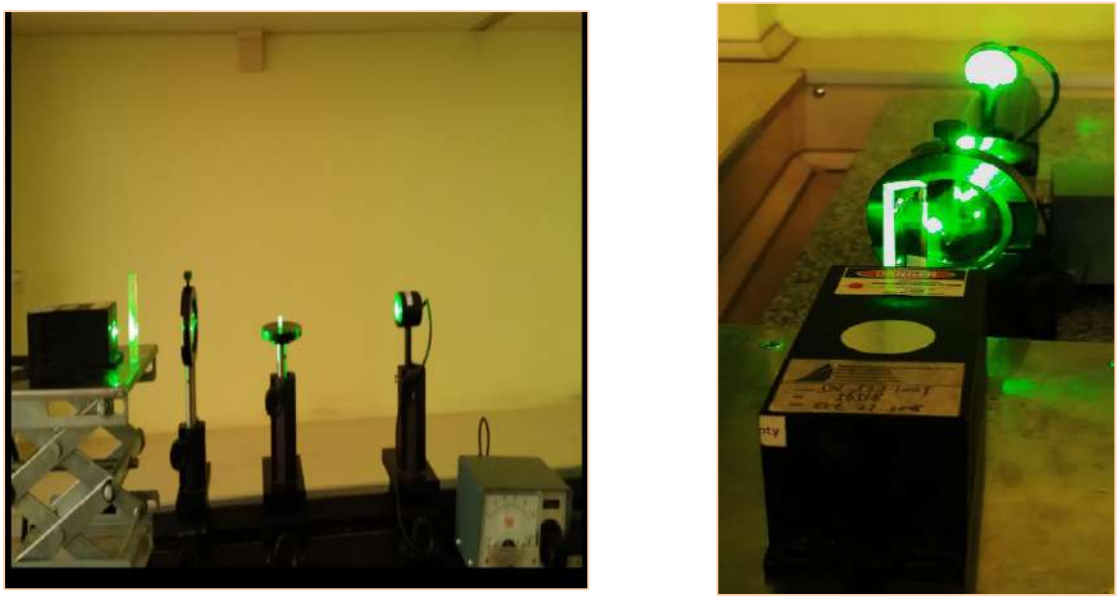


Figure (3-6) the set – up of Optical Limiting for ($KTiOPO_3$)

The laser beam focuses by lens with focal length 15 cm. The Crystal is placed at the position in focal point. A variable beam splitter (VBS) and was used to vary to input power. The input power of the laser beam is varied systematically and the corresponding output power is detected by a power meter.

3.8 Second Harmonic Generation Set –up

The figure (3-7) shows the experiment of Second Harmonic Generation for $KTiOPO_3$. The Harmonic generation output power was measured for different incident angles in the range from -35 to 35 increments of 5 step. To measure the incident angle , Bevel Protector was used as it is illustrated in figure (3-9) the incident angle of fundamental beam was changed by tilt the crystal forward to the left of optical axis to make a positive angle , and backward to the right of the optical axis to make a negative angle.

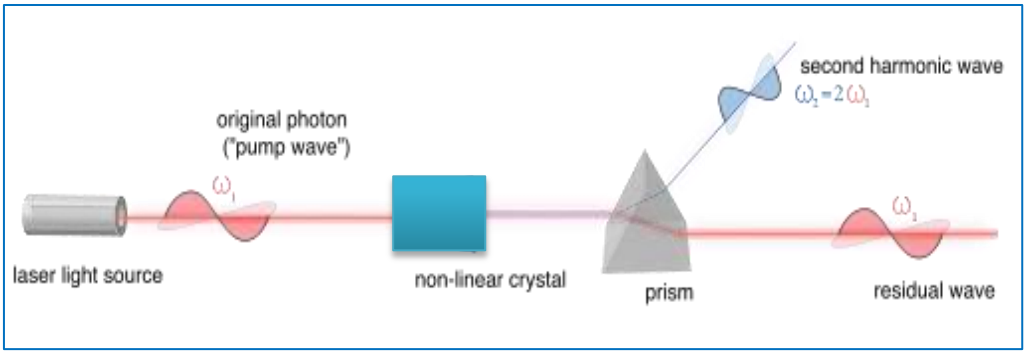


Figure (3-7) A typical configuration for second harmonic generation

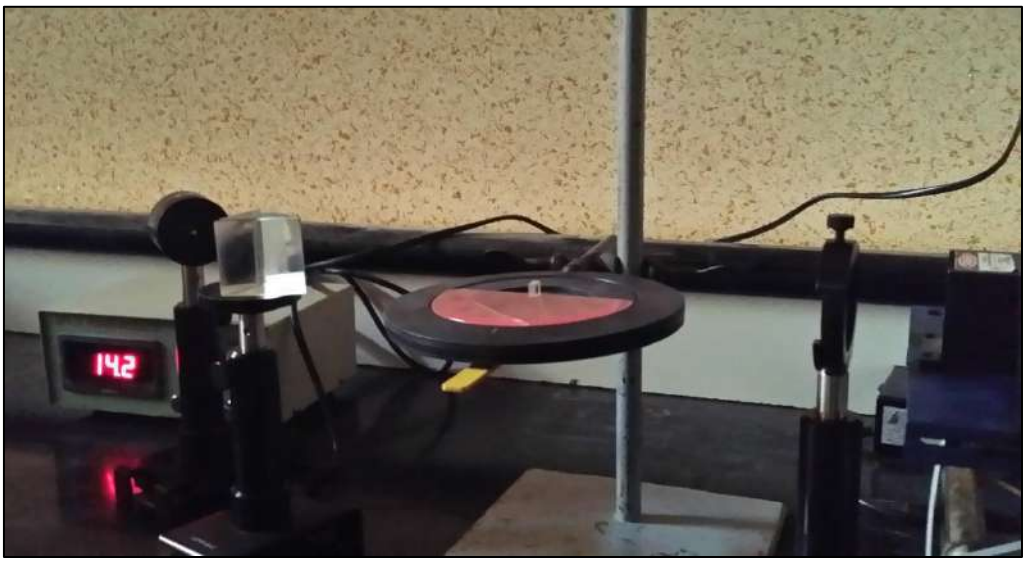


Figure (3-8) the set-up of SHG measurements at wavelength 1064nm

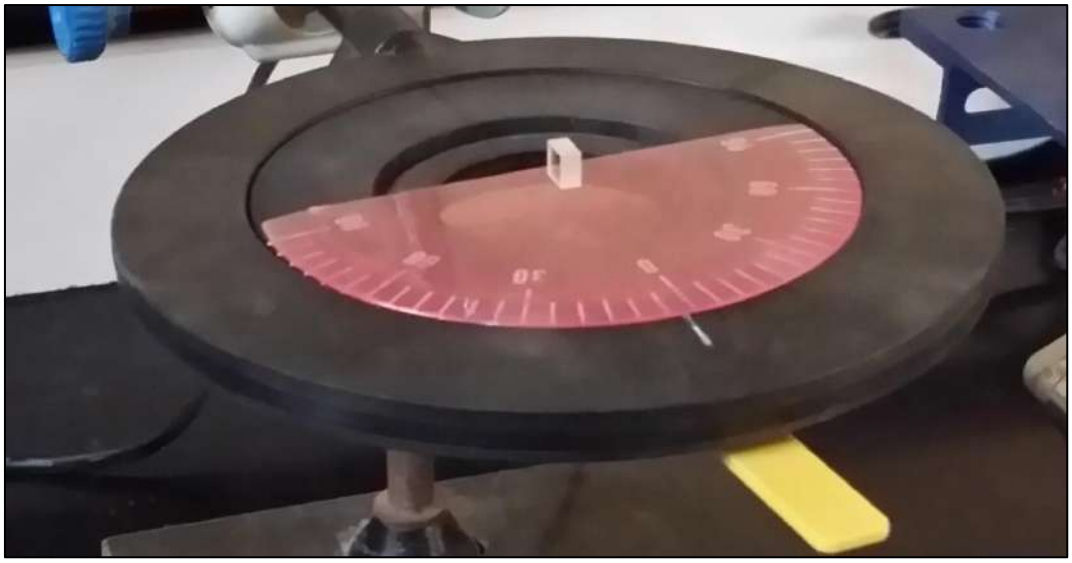


Figure (3-9) crystal thickness tilts using bevel protractor.

Table (3-3) chemical, structure, linear and nonlinear optical properties of $KTiOPO_3$ crystal [46, 80, 82, 84]

Chemical Structure	$KTiOPO_3$
Crystal Structure	Orthorhombic, space group $Pna2_1$,point group mm^2
Lattice Parameter	a=6.404 A°, b=10.616 A°, c=12.814 A°
Mohs Hardness	5
Density	3.01 g/cm^3
Linear and Nonlinear Optical Properties	
Thermal Expansion Coefficient	$\alpha_x= 11x10^{-6}/k$, $\alpha_y= 9x10^{-6}/k$, $\alpha_z=0.6x10^{-6}/k$
Transparency Range	350～4500nm
Therm-optic Coefficients (/°C)	$dn_x/dT= 6.1X10^{-6}$, $dn_y/dT= 8.3X10^{-6}$ $dn_z/dT= 14.5X10^{-6}$
Absorption Coefficients	<0.1%/cm^{-1} at 1064nm <1%/cm^{-1} at 532nm
For Type II SHG of a Nd:YAG laser at 1064nm	Temperature Acceptance: 24°C-cm^{-1} Spectral Acceptance: 0.56 nm-cm^{-1} Angular Acceptance: 14.2 mrad-cm^{-1} (φ);55.3 mrad-cm^{-1} (θ) Walk-off Angle: 0.55°
NLO Coefficients	deff(II)≈(d24-d15)sin2φsin2θ-(d15sin2φ + d24cos2φ) sinθ
Non-vanished NLO susceptibilities [pm/V]	d31=6.5 pm/V d24=7.6 pm/V d32=5 pm/V d15=6.1 pm/V d33=13.7 pm/V
Sellmeier Equations (λ in μm)	nx2=3.0065+0.03901/(λ2-0.04251)-0.01327λ 2 ny2=3.0333+0.04154/(λ 2-0.04547)-0.01408λ2 nz2=3.3134+0.05694/(λ 2-0.05658)-0.01682λ2

Table (3-4) chemical, structure, linear and nonlinear optical properties of $KTiOPO_4$ crystal

Chemical Structure	$KTiOPO_4$
Crystal Structure	Orthorhombic, space group $Pna2_1$,point group mm^2
Lattice Parameter	a=6.404, b=10.616, c=12.814?, Z=8
Density	3.024 g/cm^3
Linear and Nonlinear Optical Properties	
Thermal Expansion Coefficient	$\alpha_x=11x10^{-6}/°C$, $\alpha_y=9x10^{-6}/°C$, $\alpha_z=0.6x10^{-6}/°C$
Transparency Range	350～4500nm
Therm-optic Coefficients (/°C)	$dn_x/dT=1.1X10-5$, $dn_y/dT=1.3X10-5$ $dn_z/dT=1.6X10-5$
Absorption Coefficients	$\alpha<0.001$ (λ =1064 nm or 532 nm)
For Type II SHG of a Nd:YAG laser at 1064nm	Temperature Acceptance: 24°C-cm Spectral Acceptance: 0.56nm-cm Angular Acceptance: 14.2mrad-cm (φ);55.3mrad-cm (θ) Walk-off Angle: 0.55°
NLO Coefficients	$deff(II)\approx(d24-d15)\sin2\varphi\sin2\theta-(d15\sin2\varphi + d24\cos2\varphi)\sin\theta$
Non-vanished NLO susceptibilities	d31=6.5 pm/V d24=7.6 pm/V d32=5 pm/V d15=6.1 pm/V d33=13.7 pm/V
Sellmeier Equations (λ in μm)	$nx2=3.0065+0.03901/(\lambda2-0.04251)-0.01327\lambda 2$ $ny2=3.0333+0.04154/(\lambda 2-0.04547)-0.01408\lambda2$ $nz2=3.3134+0.05694/(\lambda 2-0.05658)-0.01682\lambda2$

CHAPTER FOUR

RESULTS

AND

DISCUSSIONN

4.1 Introduction

This chapter explains the experimental results and discussion that gained them through the study of linear and nonlinear optical properties for $KTiOPO_3$ crystal, which has dimensions of 6x6x3 mm^3. The optical results represent by absorption, transmission spectra and calculation of optical constants, the optical energy gap, and to calculate the optical conductivity.

The nonlinear optical properties represent absorption nonlinear coefficient, and refraction nonlinear index into wavelength (1064, 532) nm for Nd:YAG laser by using Z-scan technique. In addition to display results of second harmonic generation for KTP crystal in 1064nm.

4.2 Optical properties for Potassium Titanyl Phosphate

4.2.1 Linear Optical Properties

We studied the spectral of absorbance and transmittance for KTP crystal by the use of the spectrophotometer (UV-Visible). Here the obtained results from this study.

The figure (4-1) shows the absorbance spectra for KTP crystal, the highest value for the absorbance is 0.312 at wavelength 330 nm. Figure (4-2) shows the transmittance spectrum for KTP crystal, which possess highest value for transmittance, is 88.34% at wavelength (1095) nm.

Through the absorbance and transmittance spectrum, the reflective curve was calculated that is related with the absorbance and transmittance through the relation (2-4) .Also the optical constants were calculated , the linear absorption coefficient, the extinction coefficient, and the refraction index , as well as the calculation of the optical energy gap and the optical conductivity that explained in figures (4-3,4-4,4-5,4-6,4-7,4-8) receptivity.

Through our linear optical measurements for KTP crystal, we found from these measurements, the highest value for absorption coefficient is 2.399 when incident photon energy is 3.75 eV as illustrated in the figure (4-4). The highest value for refractive index is 2.621 when the incident photon energy is 3.6 eV as shown in the figure (4-5). The highest value for extinction coefficient is $6.46x10^{-9}$ when the incident photon energy 3.6 eV as illustrated in the figure (4-6). We found that the optical energy gap is 3.8 eV as shown in the figure (4-7). The highest value of the optical conductivity is 2.621 at wavelength 344 nm as illustrated in the figure (4-8).

4.2.1.1 Absorbance

The absorbance of $KTiOPO_3$ is measured by the spectrophotometer (UV-Visible) at wavelengths (300-1100) nm recorded using Mega (UV-2100) for constant thickness (3mm).

Figure (4-1) illustrates the values of absorbance as far as the wavelength that shows the $KTiOPO_3$ crystal has the highest value for the absorbance is 0.312 when incident photon energy is 330 nm. After that, rapidly it decreased with the increased of the wavelength.

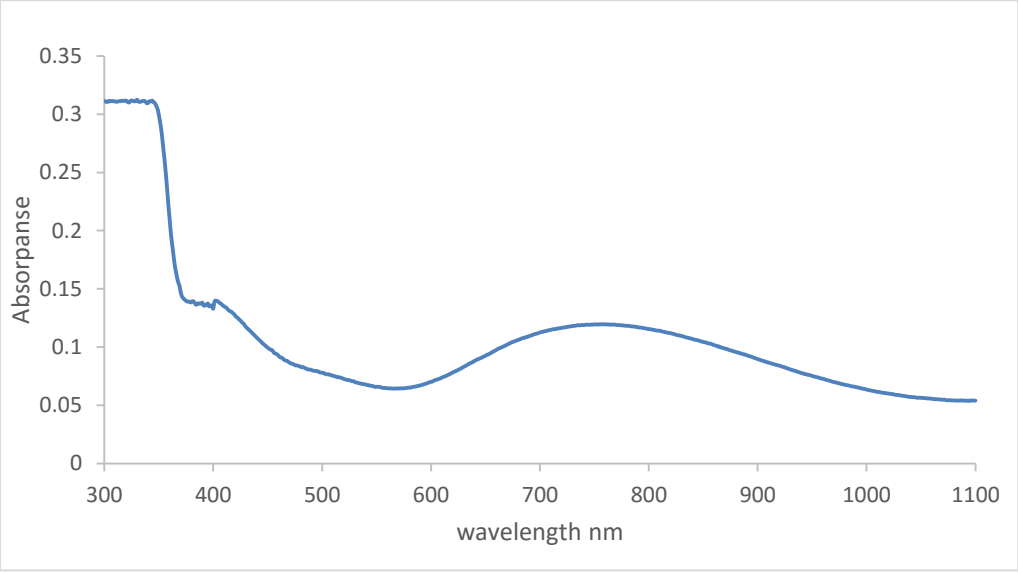


Figure (4-1): The Absorbance versus wavelength for $KTiOPO_3$.

The crystal is the strong absorption .It is caused by OH groups grown into, or trapped in the crystal during grow. The OH group oscillates as a hydroxyl molecule and forms an OH defect in the crystal. This is attribute to the vibration in the PO tetrahedral.

4.2.1.1 Transmittance

Transmittance spectra studied in wavelength range (300-1100) nm for KTiOPO3 crystal with constant thickness (3mm).The transmittance spectra depends on many factors such as quantity of energy levels, which connects in chemical structure and matter crystallization. It also depends on thickness that has a vital role in matter transmittance. Figure shows (4-2) the transmittance spectra for KTP crystal as function for wavelengths. The transmittance spectra is reversed in behavior to the absorbance spectra. It illustrates the transmittance that has a decreased with value at short wavelength then increased the transmittance with the increment of wavelength. It is observed from the same figure (4-2) the highest value for transmittance, is 88.34% at wavelength 1095 nm.

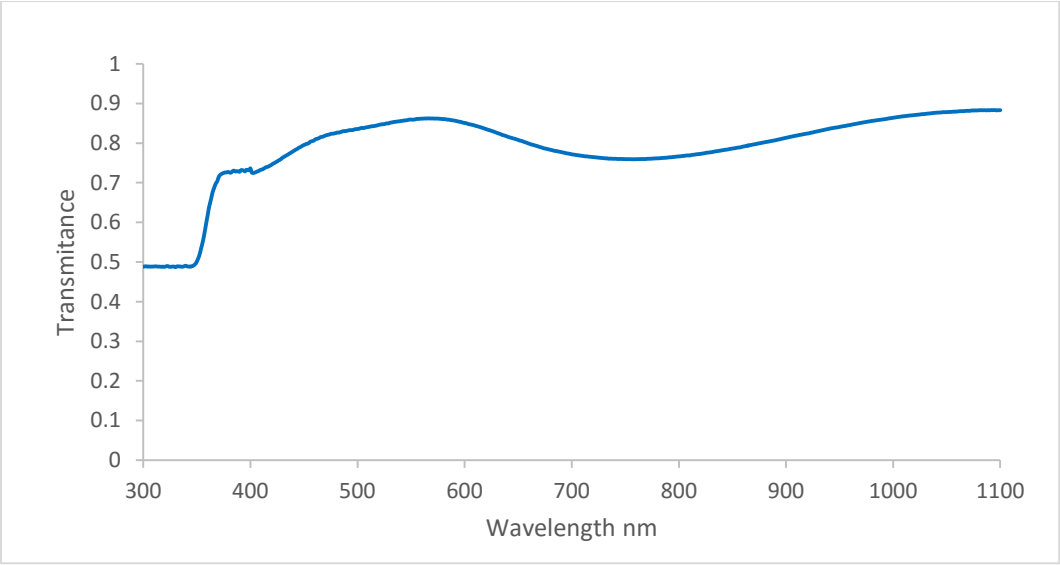


Figure (4-2): The Transmittance versus the wavelength.

4.2.1.3 Reflection

The calculation of reflection depending on absorption and transmittance by the relation (2-4). The figure (4-3) displays the curve of reflection as a function of photon energy for KTP crystal and shows the similarities of this curve with absorption coefficient because reflection conect in a relation with absorbance. The highest value begins for reflective spectra is 0.2 when incident photon energy is 3.5 eV and up.

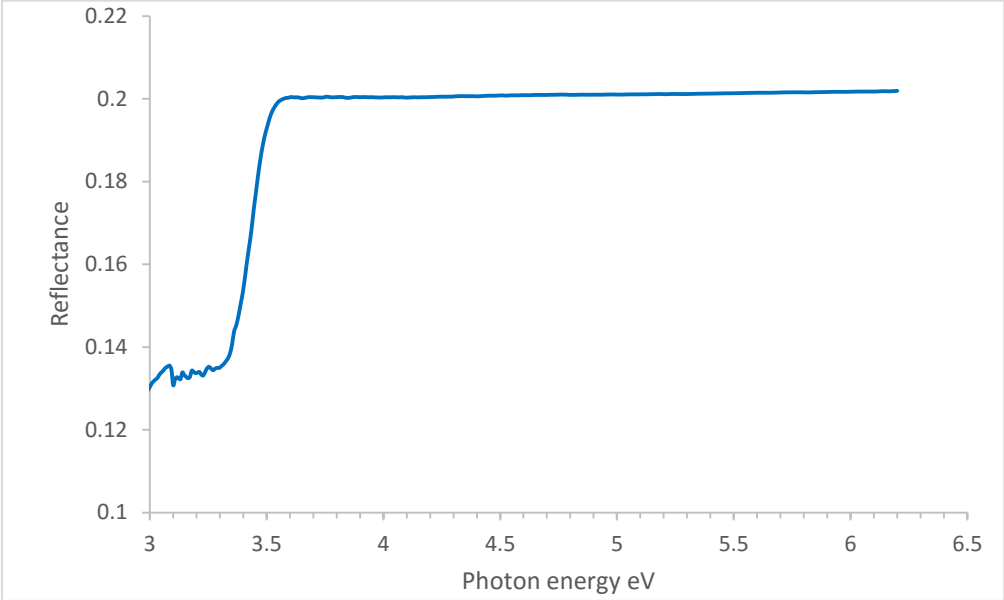


Figure (4-3): Reflectance spectra for $KTiOPO_3$

4.2.1.4 Absorption Coefficient

The calculation of absorption for KTP crystal in thickness 3mm through the relation (2-6). The absorption coefficient depends on incident photons energy and on the properties of the matter that represented by energy gap and electronic transmission types that happened among energy bands. The figure (4-4) shows the relation of absorption coefficient as a function for photon energy for KTP crystal, the values of absorption coefficient increased gradually with the increase of photon energy then it reaches highest value for absorption coefficient 2.399 when incident photon energy is 3.75 eV and up.

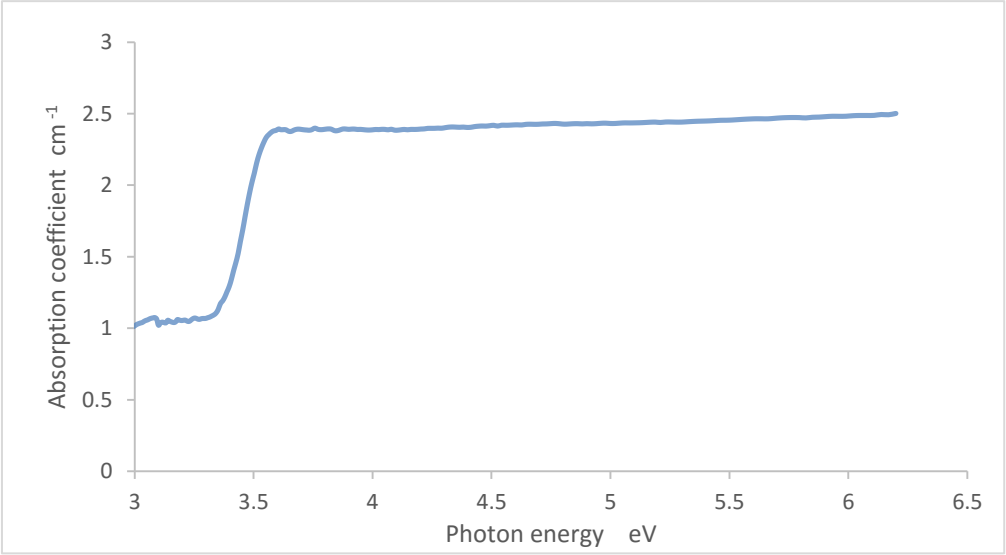


Figure (4-4): Absorption coefficient versus photon energy.

4.2.1.5 Refraction Index

Through the relationship (2-7), the refraction index has been calculated of KTP crystal. The figure (4-5) explains the change of refraction index with photon energy, and the highest value for refractive index is 2.621 when the incident photon energy is 3.6 eV. Therefore, it is observed that the refraction curve similar to reflection curve because of the connection the reflection with the refraction index according to relation (2-7).The refraction index depends on the type of material and its crystallization structure.

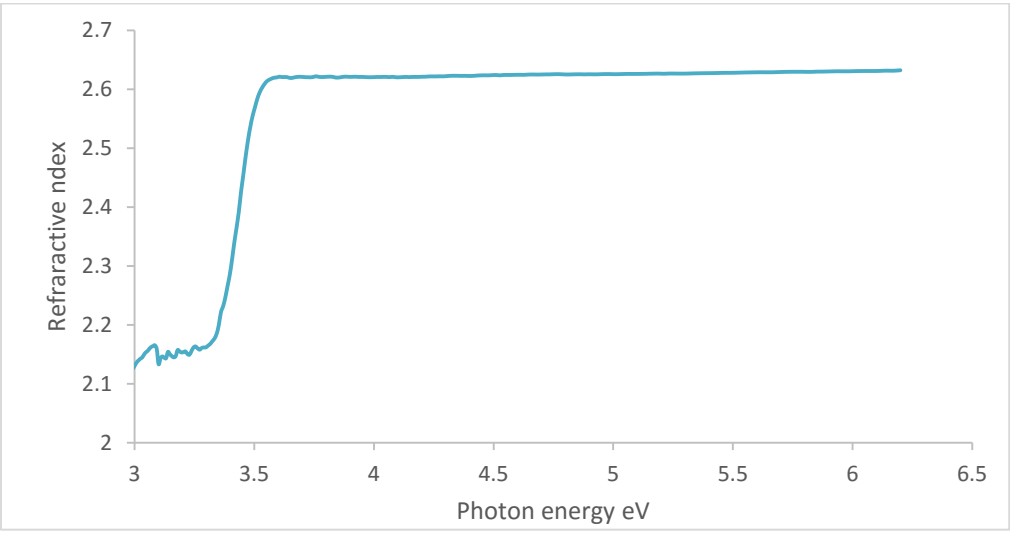


Figure (4-5): Refractive index variation of photon energy

4.2.1.6 Extinction coefficient

Extinction coefficient was calculated through the values of absorption coefficient by using the equation (2- 8). The figure (4-6) shows the change of extinction coefficient with photon energy for KTP crystal and the highest value for extinction coefficient is 6.32×10^{-5} when the incident photon energy 3.67 eV.

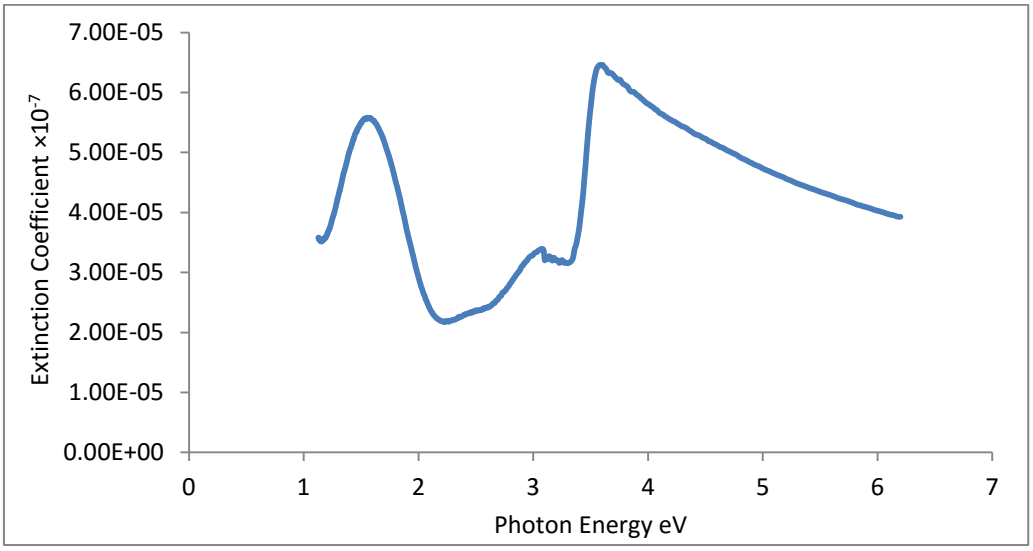


Figure (4-6): Extinction coefficient versus incident photon energy.

4.2.1.7 The Band Energy Gap

The value of optical energy gap is one of the most important of optical magnitudes that effected by the structure of material. The energy gap of $KTiOPO_3$ crystal determined for forbidden direct electronic transition by using the relation (2-9), it was r=3/2. The draws the relation between (hν) 3/2 and photon energy (hν) and draws the straight part to cut axis of photon energy at the point (hν) = 0. The energy gap to forbidden direct transition , is (3.8eV) for KTP crystal as shown in figure (4-7).This magnitude is near for $KTiOPO_3$ energy value when our comparison for this results with the results of energy gap to $KTiOPO_4$ crystal that found in[],[] and the comparison was corresponding.

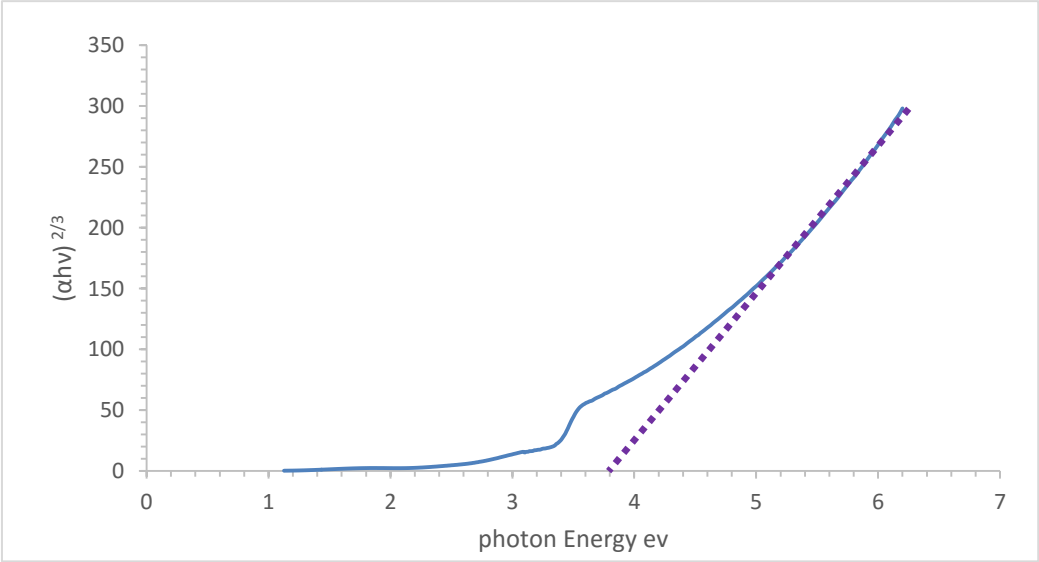


Figure (4-7): The variation of the $(\alpha h\nu)^{2/3}$ with the incident photon energy.

4.2.1.8 Optical Conductivity

The optical conductivity of KTP crystal is as shown in figure (4-8) .The value of this optical conductivity is calculated from the equation (2-10).From this figure, the highest value of the optical conductivity is (1.44×10^{11}) at the incident photon energy 3.5eV and up, and it is clear that the conductivity as function for photon energy of $KTiOPO_3$ crystal. Addition that it is absorption coefficient from the same figure that the curve of the optical conductivity is similar to the absorbance which combined with the equation (2-10).

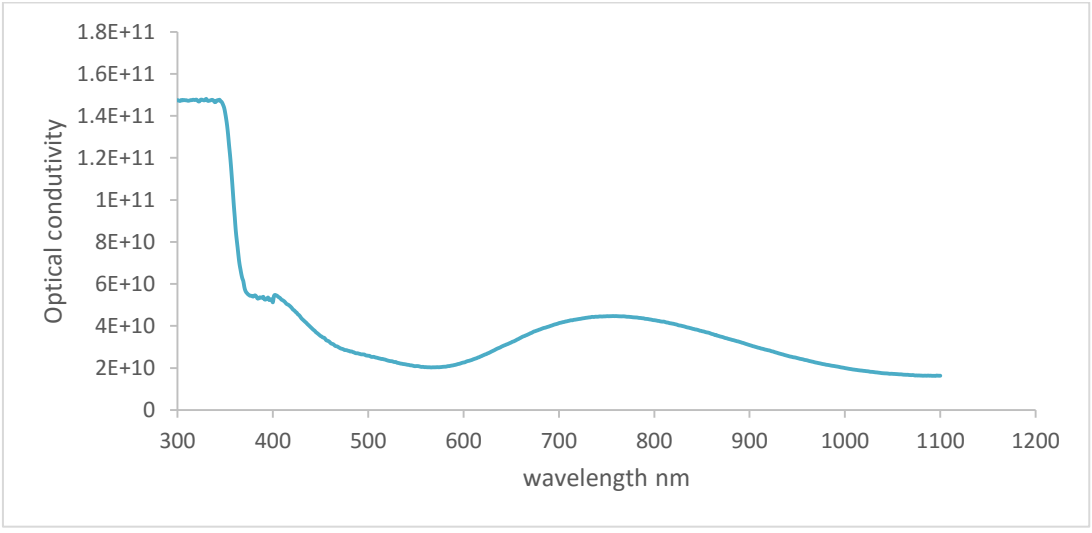


Figure (4- 8): The optical conductivity versus the wavelength.

The result of linear optical properties for Potassium Titanyl Phosphate ($KTiOPO_3$ or KTP) crystal illustrated in the Table (4-1).

Table (4-1) linear optical properties for Potassium Titanyl Phosphate ($KTiOPO_3$ or KTP).

λ(nm)	t (cm)	T%	$\alpha(cm)^{-1}$	$K*10^{-5}$	R	n
1064	0.3	0.88	0.42375	3.539	0.064	1.67
532	0.3	o.85	0.5335	2.228	0.078	1.77

4.2.2 Linear Optical Properties Theoretically

So that to reproved our Potassium Titanyl Phosphate ($KTiOPO_3$) crystal model results ,the reliance of the refractive index on the wavelength and the suitable range of angles incident for the reflectance in order to phase matching condition .Through the programming on the international data (Refractive index database) are applied as sketched in the figure (4-9) and (4-10)respectively.

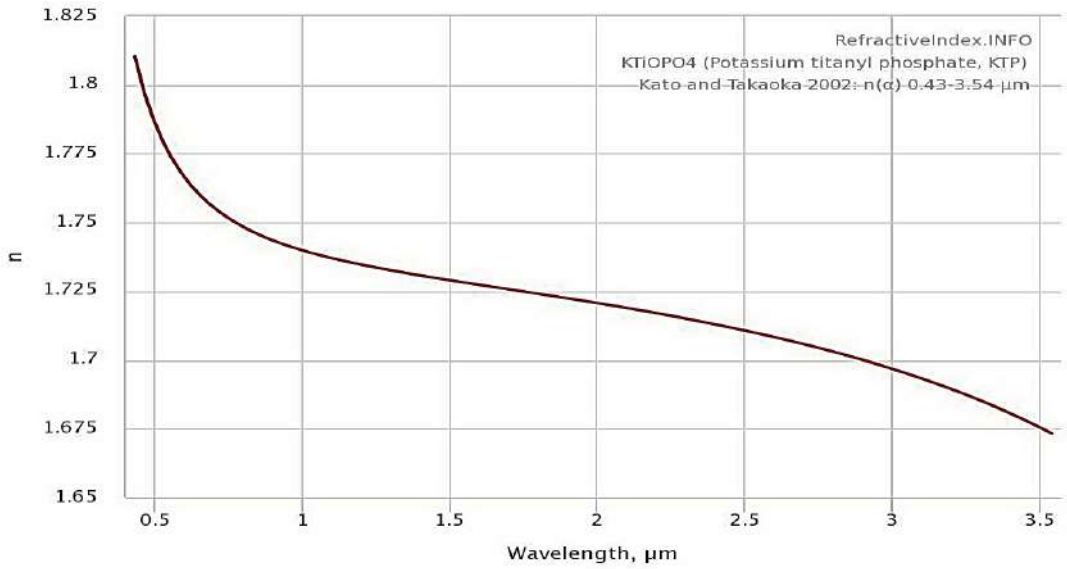


Figure (4-9): The refractive index on the wavelength (https// refractive index.info)

The figure (4-9) shows that refractive index of $KTiOPO_3$ decreases gradually with the increased of the wavelength .The refractive index at 1064nm was (1.67) and (1.77) at wavelength 532nm, that is agreement with our experimental results.

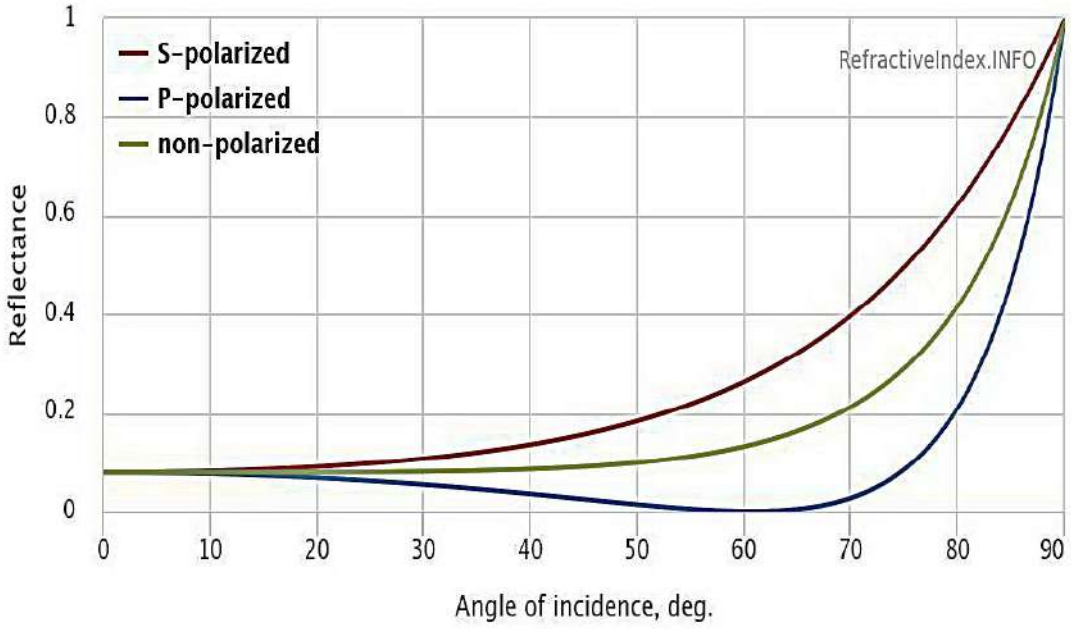


Figure (4-10) Dependence of reflectance on the angle of incident (https// refractive index.info)

In the figure (4-10) is illustrated the relation between the reflectance and the incident face of KTP crystal, and noticed that the reflectance from the incident face of KTP crystal is 35° degree.

4.2.3 Nonlinear optical properties

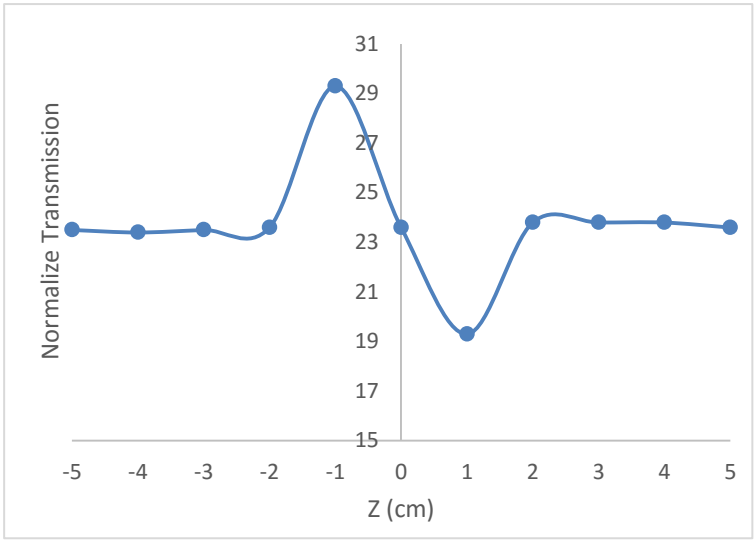
The practical results that we obtained from Z-scan technique for both close and open aperture to Potassium Titanyl Phosphate KTP crystal by using laser CW Nd:YAG in to two wavelengths 1064 nm and frequency doubler 532 nm with different powers(25,35,80) mW as illustrated in the figures(4-11,4-12, and 4-13) respectively.

The magnitude of nonlinear optical coefficients for Potassium Titanyl Phosphate KTP crystal has been calculated. The calculation of nonlinear refraction index (n_2) value from the equation (2-29), the change in the value of refraction index (Δn) from the equation (2-30), whereas the magnitude of nonlinear absorption coefficient calculated from the equation (2-31), as well as the absolute value the third order nonlinear optical susceptibility $|\chi^{(3)}|$from the equation (2-32).

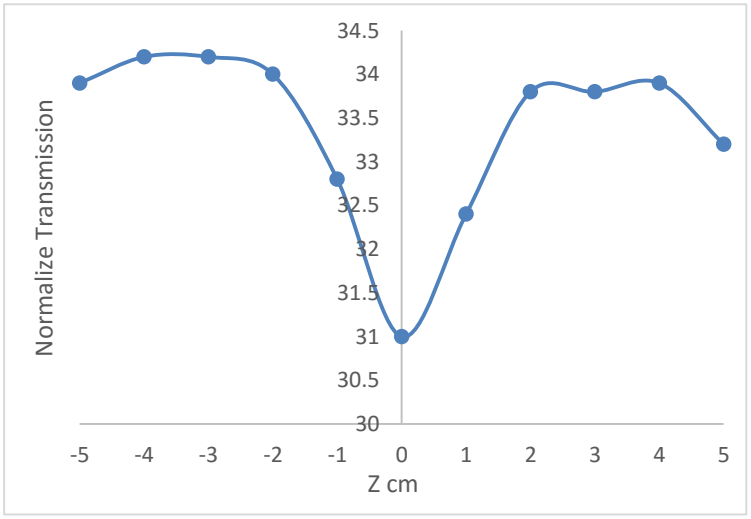
The figures (4-11 (a),4-12 (a), 4-13 (a)) represent the transmittance curve of KTP crystal depends on a different position in the close aperture Z-scan, and the figures (4-11 (b), 4-12 (b), 4-13 (b)) The transmission curve of KTP crystal depends on a different position in the open aperture Z-scan.

To obtain the nonlinear refraction index only, the values of transmission laser beam in case of close aperture is divided on the values transmission in case of open aperture as illustrated in the figures (4-11 (c), 4-12 (c), 4-13(c)).

(a)



(b)



(c)

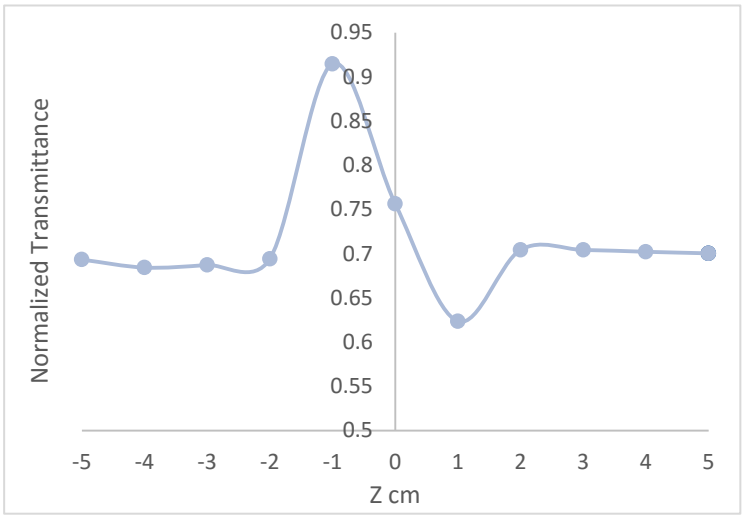
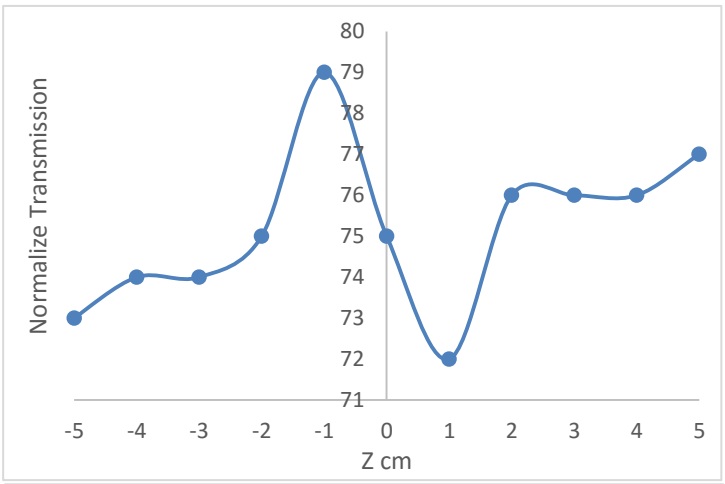
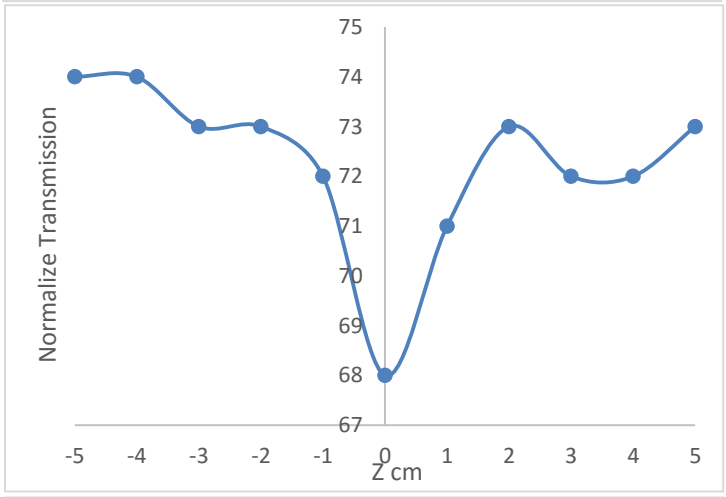


Figure (4-11): The normalize transmittance curve as a function of position for the crystal at wavelength 1064nm with power 35mW. (a) The transmittance curve when the close aperture (b) when the open aperture. (c) The transmittance curves that resulted from the division of the transmittance curve for the close aperture to transmittance curve for open.

(a)



(b)



(c)

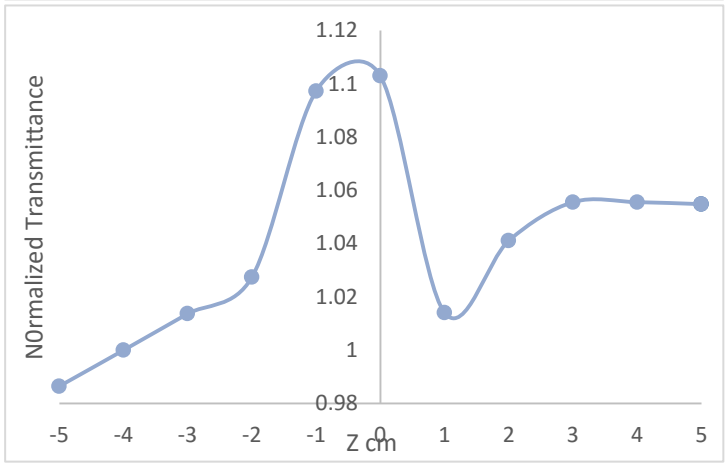
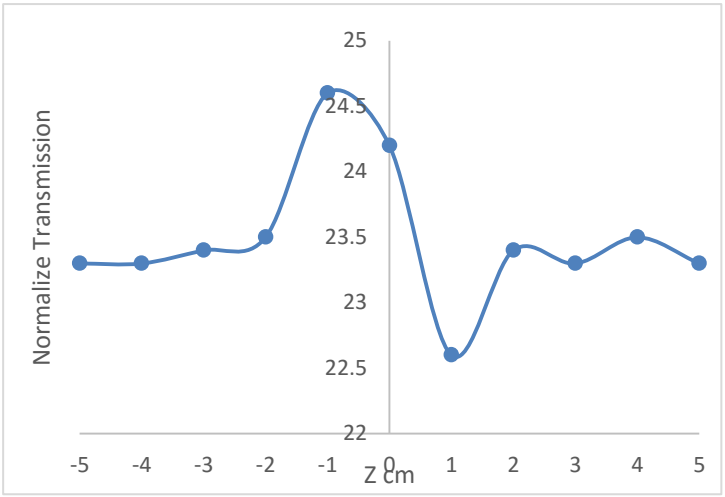
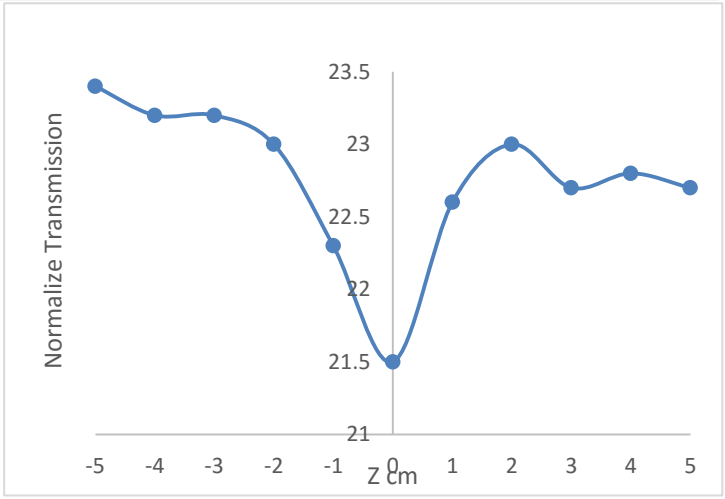


Figure (4-12): The normalize transmittance curve as a function of position for the crystal at wavelength 532nm with power 80mW. (a) The transmittance curve when the close aperture (b) The transmittance curve when the open aperture. (c) The transmittance curves that resulted from the division of the transmittance curve for the close aperture to transmittance curve for open.

(a)



(b)



(c)

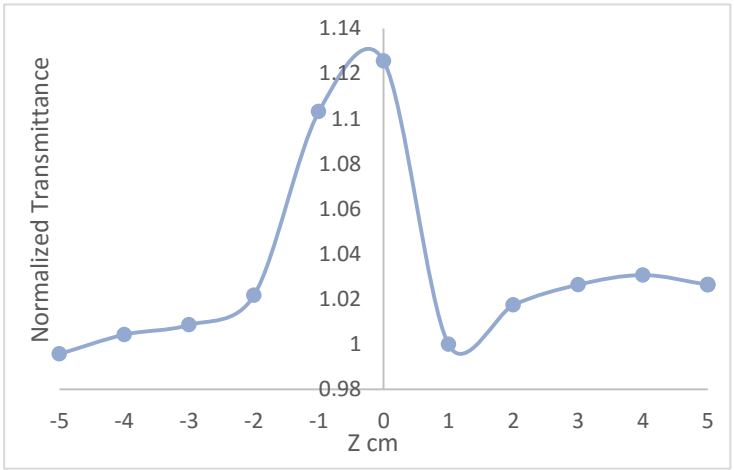


Figure (4-13): The normalize transmittance curve as a function of position for the crystal at wavelength 532nm with power 25mW. (a) The transmittance curve when the close aperture (b) The transmittance curve when the open aperture. (c) The transmittance curves that resulted transmittance curve for the close aperture to transmittance curve for open.

The observed from the results of Z-scan technique for KTP crystal that the nonlinear refraction index value is $3.59x10^{-14}$ cm^2/mW at wavelength 1064 nm, and the value of nonlinear refraction index n_2 is $3.67x10^{-14}$, $3.35x10^{-14}$ cm^2/mW at the wavelength frequency doubler 532 nm with powers 80, 25 mW receptivity.

The magnitude of nonlinear absorption coefficient β is $2.71x10^{-3}$ cm/mW at wavelength 1064 nm. The magnitude of nonlinear absorption coefficient β is $4.32x10^{-3}$, $3.37x\ 10^{-3}$ cm/mW at wavelength frequency doubler 532nm with two powers 80, 25 mW receptivity.

The value of the third order nonlinear optical susceptibility $|\chi^{(3)}|$ is $2.55x10^{-5}$ at wavelength 1064 nm with power 35 mW. Its value is $2.28x10^{-5}$, $2.32x10^{-5}$ at wavelength frequency doubler 532 nm with two powers 80, 25 mW receptivity.

The Table (4-2) is summarized the nonlinear optical properties that calculated for KTP crystal.

Table (4-2): The results of nonlinear optical properties for $KTiOPO_3$ crystal by Z-scan technique.

λ (nm)	Power of laser(mW)	ΔΦ (Rad)	ΔT_{p-v}	$n_2\times10^{-14}$ (cm^2/mW)	Δn × 10^{-9}	T_{min}	β $\times10^{-3}$ (cm/mW)	$\lvert\chi^{(3)}\rvert\times10^{-3}$ (esu)
1064	35	24.137	9.8	3.59	4.08	31.2	2.71	2.55
532	80	17.241	7	3.67	5.77	68	4.32	2.28
	25	4.926	2	3.35	1.65	21.5	4.37	2.32

Moreover, we found that the values of nonlinear absorptions coefficient influence by the wavelength. The results of our study of KTP crystal, that the nonlinear absorption coefficient β $4.32x10^{-3}$, $4.37x10^{-3}$ cm/mW at the wavelength frequency doubler 532nm, with for two different intensities larger

than the magnitude of that nonlinear absorption coefficient 2.71x 10^{-3} at wavelength 1064 nm. As illustrated in figure (4-14), because the absorbance of crystal decrease with the increase of wavelength.

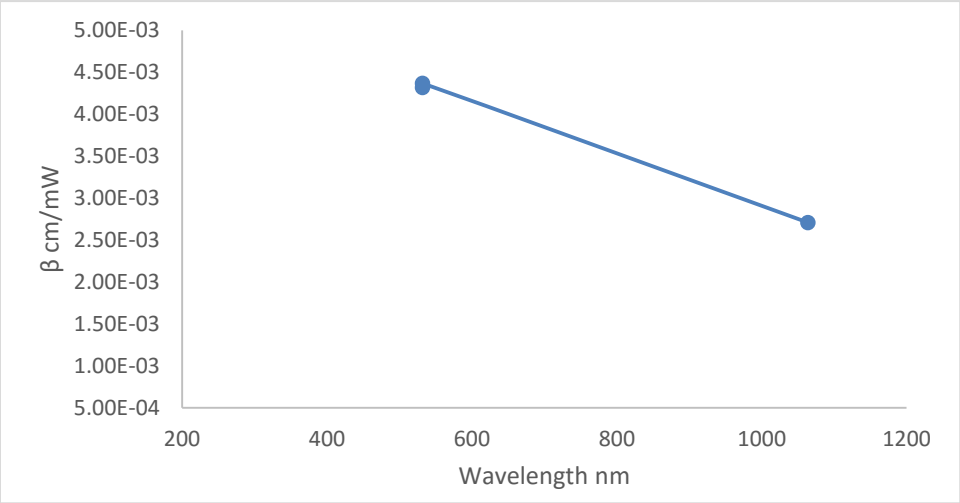


Figure (4-14) nonlinear absorption coefficient a function to the wavelength

From the result that obtained for Potassium Titanyl Phosphate KTP crystal from the Z-scan technique for both close and open aperture , we found that KTP crystal has self-defocusing effect and two photons absorption , also it has a high values to the third order nonlinear optical susceptibility $\chi^{(3)}$.

4.3 Optical Power Limiting

We made a study to optical power limiter for Potassium Titanyl Phosphate KTP crystal by use Z-scan technique and Nd: YAG laser CW at frequency doubler 532 nm with power rang (10-85) mW. The figure (4-15) represent the optical power limiter. It is observed from this figure that KTP crystal occurs a good optical power limiter at 60 mW threshold.

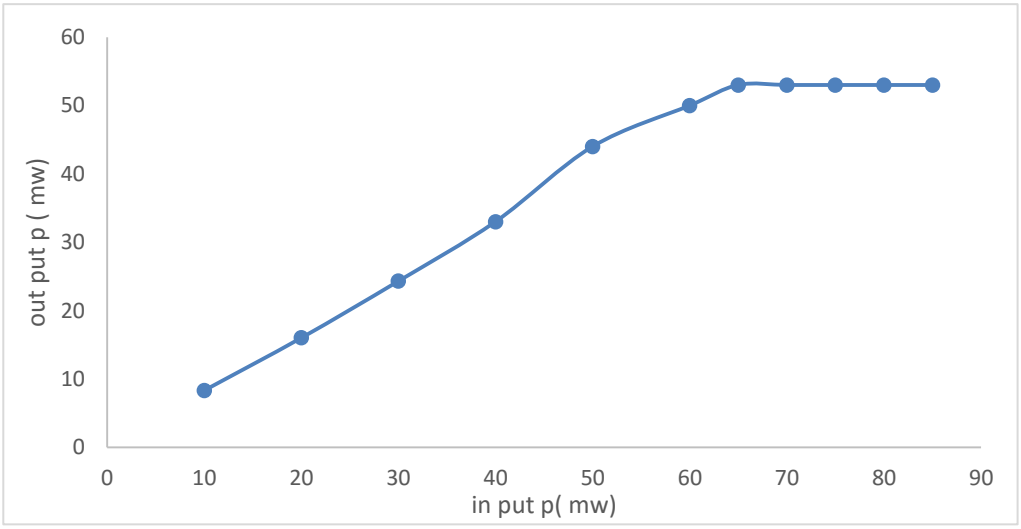


Figure (4-15) The Optical Limiting for KTP crystal.

The figure (4-15)shows the input power in the range (10,20,30,40, 50 ,60,65,70,75,80,85) mW .the output beam power increases with increasing input beam power for KTP crystal ,up to 60mw the output beam power is constant, because its nonlinear absorption coefficient increases with increases in the incident irradiance. The volume of limiting threshold for KTP crystal is found 60 mw. Result that KTP crystal is good for optical limiter. To obtain other representation for optical power limiter, we draw transmittance laser beam (output power/ input power) on the expense input laser beam power to crystal, as it is shown in the following figure (4-16).

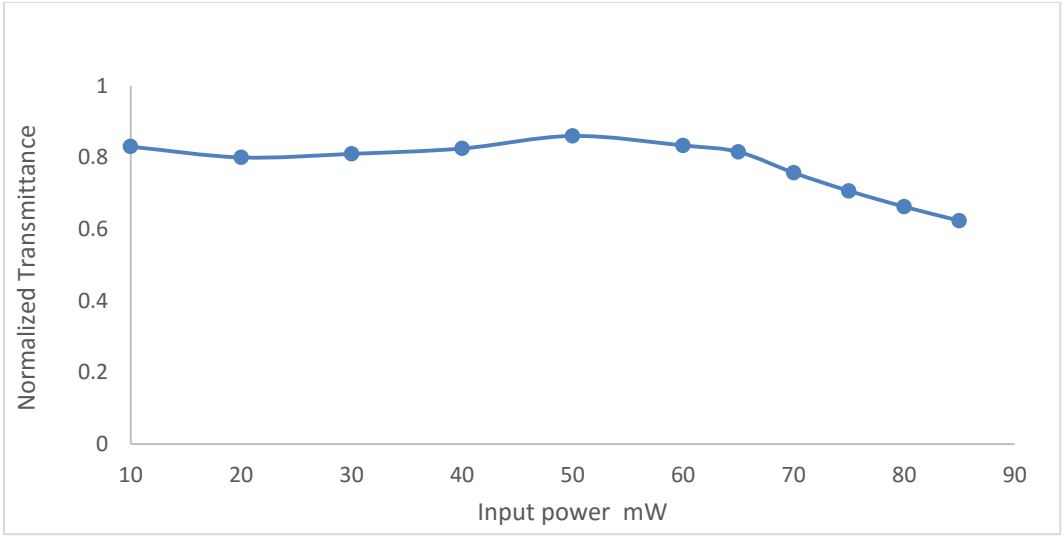


Figure (4-16): The transmittance curve via input power for KTP crystal.

The values of optical limiter for $KTiOPO_3$ crystal listed in Table (4-3).

Table (4-3) the value of the power-limiting threshold.

crystal	λ (nm)	Power Limiting Threshold L_m (mw)
KTP	532	60

4.4 The Intensity of Second Harmonic Generation.

The figure (4- 17) shows the experimental results. The intensity of harmonic generation as a function of the incident angle range which was obtained at -35° and 35°. The path length inside the KTP crystal was decreased when increasing the incident angle .Harmonics generation intensity was disappear at the critical incident angles because no signal will be detected at the critical angles.

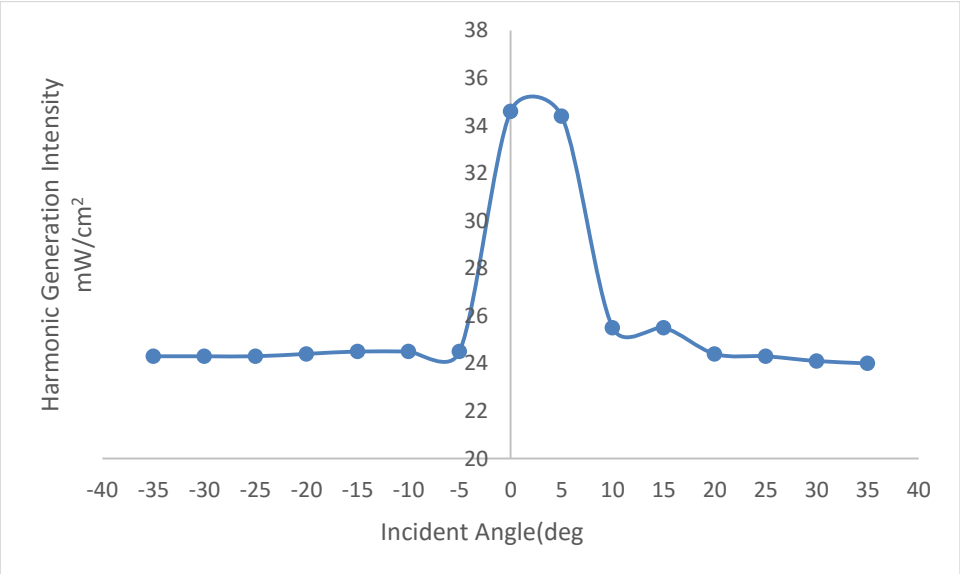


Figure (4-17) Harmonic generation output intensity versus incident angle

The different values of harmonic generation of intensity at each the input power due to the charge in the incident angle of fundamental beam. The cause of the change in the incident angle of the fundamental beam is to get the efficient

harmonics generation intensity at was indicated a change in refractive index, which is well agreement with our theoretical results, illustrated figure (4-9).

When the input power of 1064nm Nd: YAG laser was 35mw the generated of the second harmonics 532nm by using KTP crystal was obtained at incident angle 1^{o} with 14.3mW powerful.

The efficiency of harmonic generation can be calculated from equation (2-16). It can be noticed from the results in Table (4-4).

Table (4-4) present the result experimental of Second Harmonic Generation of Potassium Titanyl Phosphate (KTiOPO3 or KTP).

Laser Type	Input Power (mW)	Harmonic Generation	The efficiency %
Nd:YAG 1064nm	35	SHG	13

4.5 Conclusion

Through the results that obtained from the study of optical properties, the nonlinear optical properties, optical power limiting and second harmonic generation for KTP crystal conclude the following:

1- KTP crystal has a high transmittance because of low of attenuation. The calculation of energy gap has been determined for direct forbidden uses; it is found that it equals (3.8 eV).

2- The results presented the simplicity, sensitivity and cases Z-scan technique and determined the nonlinear refraction index , nonlinear absorption coefficient, Nonlinear optics susceptibility $\chi^{(3)}$ and also the change in refraction index Δn , KTP crystal shows self-defocusing and two photons absorption, moreover it shows that KTP crystal has high values for both nonlinear refraction index and nonlinear absorption coefficient ; good and acceptable results are achieved between theoretical and practical. The crystal has large nonlinear properties.

3-The limiting effect of the crystal occurred at a threshold power value of 60mw for 532nm and measured from deviation of linearity.

4- KTP crystal possesses a high linear properties and a large response to the nonlinear effects, which enables this crystal to be fit material at the linear, nonlinear, optical power limiter and second harmonic generation applications.

5- The result of the second harmonic generation that the crystal is excellent for second SHG to convert the ultraviolet frequency to the visible.

4.6 Future Works

In the program of our work, .We suggest the following:

1-Study on the performance of $KTiOPO_3$ crystal as an optical limiter with different laser wavelength.

2-Investigation of KTP crystal activation by using z- scan technique for two laser beams.

3-The related experiments by the generation of third and higher for KTP crystal.

4-Invistigate on using KTP crystal to combination the optical parametric oscillator of type II phase matching, with the second harmonics generation.

5-The study of the better performance of KTP crystal as an element for Q-Switching solid –state laser with high power pulsing.

6-The study the nonlinear optical properties of KTP crystal to motivate the three wave mixing.

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الخلاصة

الدراسة الحالية تتضمن دراسة الخصائص البصرية الخطية و اللاخطية وتأثير محدد القدرة البصري والتوليد التوافقي الثاني لبلورة بوتاسيوم تيتانيل فوسفات (KTP) كعنصر فعال والتي ابعادها (3 × 6 × 6) ملم^3 باستخدام ليزر النديميوم –ياك الذي يعمل بالموجة المستمرة (Continuous wave CW) عند الطول الموجي 1064 نانومتر بقدرة 35ملي واط والتردد المضاعف 532 نانومتر وبقدرتين 80و 25ملي واط على التوالي.

أطياف الامتصاصية والنفاذية لبلورة بوتاسيوم تيتانيل فوسفات، تم تحليلها باستخدام جهاز المطياف ((Spectrophotometer وتبين ان بلورة KTP تمتلك نفاذية عالية تزداد بزيادة الطول الموجي وان اعلى نفاذية للبلورة تكون 88% عند الطول الموجي 1095 نانومتر، بسبب نقصان التوهين، وقد حددت فجوة الطاقة البصرية للانتقالات المباشرة الممنوعة للبلورة وتكون 3.8 الكترون فولط.

درست الخصائص البصرية اللاخطية لبلورة باستخدام تقنية مسح حزمة الليزر على المحور Z وليزر النديميوم -ياك Nd: YAG الذي يعمل بالموجة المستمرة (Continuous wave CW) عند الطول الموجي 1064 نانومتر بقدرة 35ملي واط والتردد المضاعف 532 نانومتر وبقدرتين 80و 25ملي واط على التوالي.

تم حساب معامل الانكسار اللاخطي من خلال طريقة الفتحة المغلقة (المغلقة جزئيا) وكانت قيمته 3.59 × 10^{-14} سم^2 / ملي واط و 3.35 × 10^{-14} و 3.67 ×10^{-14}سم^2 /ملي واط . اما معامل الامتصاص اللاخطي فقد تم حسابه من خلال طريقة الفتحة المفتوحة وكانت قيمته 2.71×10^{-3} سم /ملي واط 4.32 ×10^{-3} و 4.32 ×10^{-3}سم /ملي واط .

أظهرت النتائج التي تم الحصول عليها ان البلورة تمتلك معامل انكسار ذو قيمة سالبة وحدوث تأثير عدم التركيز الذاتي (Self-defocusing) و تمتلك معامل امتصاص (β) لا خطي ذو امتصاص فوتونين. كما تم حساب التغير في معامل الانكسار وحساب القيمة المطلقة للتأثيرية البصرية اللاخطية.

كذلك درس محدد القدرة البصري (The Optical Power Limiting) لبلورة KTP باستخدام تقنية مسح لحزمة الليزر Z ولقد حصلنا على خواص جيدة لمحدد القدرة البصري لبلورة KTP. تم إيجاد عتبة محدد القدرة البصري والتي تساوي60 ملي واط.

النتائج التي تم الحصول عليها تبين ان بلورة KTP تظهر تأثيرات لاخطية كبيرة مما يجعلها لان تكون مادة جيدة في تطبيقات الأجهزة البصرية اللاخطية ومحدد القدرة.

كما درست القدرة على توليد التوافقية الثانية لليزر النديميوم – ياك (Nd: YAG) ذو الموجة المستمرة CW وبقدرة 35 ملي واط . وكانت كفاءة التحويل لتوليد الطول الموجي الاخضر 13% . ان نموذج بلورة بوتاسيوم تيتانيل فوسفات KTP كانت محققة جدا كنموذج للظواهر اللاخطية.

جمـــــهورية الـــــــــعراق
وزارة التعليم العالي والبحث العلمي
جامـــــعة القـــــــادسية
كلية التربية
قسم الفيزياء

البحث في أداء KTP كعنصر فعال للحد البصري والتوليد التوافقي لليزرات المرئية وتحت الحمراء

رسالة مقدمة

الى

عمادة كلية التربية- جامعة القادسية

وهي جزء من متطلبات نيل شهادة ماجستير في علوم الفيزياء

من قبل الطالبة

منار ليلو دايخ

بكالوريوس في الفيزياء

بأشراف

أ. د رعد شاكر عبيس النائلي

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