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Acceptance Letter

18 November 2017

Dear Author(s)

Dr./Mr. Raad S. Alnaily/ University of AL-Qadisiyah, IRAQ

Mrs. Manar L. Dayekh/ University of AL-Qadisiyah, IRAQ

Greetings, With reference to your article (UOQCS 7) entitled:

Study Second Harmonic Generation of KTP Crystal

We wish to bring to your kind notice the following

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BIOTECHNOLOGY)

Study Second Harmonic Generation of KTP Crystal

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Abstract: In this paper, we tried to investigate to display the performance of the pure single potassium Titanyl Phosphate (KTiOPO₃ or KTP) crystal of dimension (6×6×3) mm³ as a distinguished proposal model of second harmonic generation and using CW Nd:YAG laser that has the spectral wavelength 1064nm to generate 532nm . The efficiency of the frequency doubler for Nd:YAG laser by means of this proposed crystal element were satisfied with both arrangements. They was 13% one step. The nonlinear optical properties as third order, such as the nonlinear refractive index and nonlinear absorption were also measured in this work to indicate on the high nonlinearity optics of these electro-optical material with the infrared and visible wavelengths range.

Kay Words: - Second harmonic generation, second-order susceptibility $\chi^{(2)}$, potassium Titanyl phosphate (KTiOPO₃ or KTP), Nonlinear refraction index , Nonlinear absorption coefficient, Third order nonlinear optical susceptibility $\chi^{(3)}$.

Introduction:

One of the most important nonlinear optical processes for technical applications is the generation of harmonics from laser light [D. Kornack, 2001]. Second harmonic generation is a nonlinear optical process [I. Rau and F. Kajzar, 2008]. Second harmonic generation (SHG) is also called frequency doubling was first demonstrated by Franken et al in 1961 [R. Sh. Alnayli, 2008]. And extended to difference materials and applications [R. Sh. Alnayli, 2008]. To approach the ideal conditions to optimized the performance of the single pure Lithium Triborate crystal (LBO) (The boron-oxygen compound LiB₃O₅ as a Nonlinear Optical crystal). as proposal element model for the combination of the Second Harmonics Generation (SHG) and Third Harmonic with fundamental Nd:YAG laser 1.064 μ m and its frequency doubling 533 μ m Nd has been

treated. [Z. S. Shanon, et al, 2016] [Z. S. Shanon et al, 2016]. The Optimum cut transversal KDP Q-switches for Nd:YAG lasers has been investigated by R. Sh. Alnayli et al.

In this paper, we described the experimental results obtained on the second-order nonlinear optics response of Potassium Titanyl Phosphate KTiOPO₃ crystal. The purpose of the present work is to study the second-order response of the Potassium Titanyl Phosphate crystal.

KTP was first developed in the end of the 1970 [J. Hellstrom, 2001]. It got a new attention. Tordjman et al analyzed the crystal structure in details in 1974. Dupont Inc, USA, started to investigate KTP's nonlinear optical as well as its mechanical properties in 1976 [S. Wang, 2005]. The KTP is biaxial crystal [D. N. Nikogosyan, 2003]. The crystal possesses a

high nonlinear, it has a high resistant to optical and mechanical damage, and its transparency range extent from the ultraviolet to the end of the mid-infrared part of the spectrum[5]. KTP has been widely used in various nonlinear optical applications, in particular in the second harmonic generation SHG and optical parametric oscillator OPO devise based on pumping with 1 μ m radiation from Nd lasers [H.Karsson,1999].KTP crystal is orthorhombic and belong to the acentric point group mm2 [4]. For this point group symmetry class, there are five non-zero nonlinear coefficients, d15 = 6.1 pm/V, d31= 6.5 pm/V, d24= 7.6 pm/V, d32= 5.0 pm/V, and d33= 13.7 pm/V [R.W.Munn,1993] [M. J. Weber,2003] .These values are still considerably higher than for many other nonlinear materials including, β -BaB₂O₄ and LiB₃O₅ [R.W.Munn,1993]. Potassium Titanyl Phosphate (KTP) crystal is nonlinear crystal .The famous type is KTiOPO₄.Another type such as KTiOPO₇, KTiOPO₃.All these types are Orthorhombic and belonged to it with only slightly different lattice parameters.

Potassium Titanyl Phosphate (KTiOPO₃ or KTP) is the most commonly used material for frequency doubling of Nd:YAG and other Nd: doped lasers, It plays an important role for parametric sources for tenable outputs from visible (600nm) to mid-IR. It is widely used in both commercial and military lasers including laboratory and medical systems, range finders, lidar, optical communication an industrial systems[thatshigh.com]. Figure (1) shows the KTiOPO₃ (KTP) crystal.

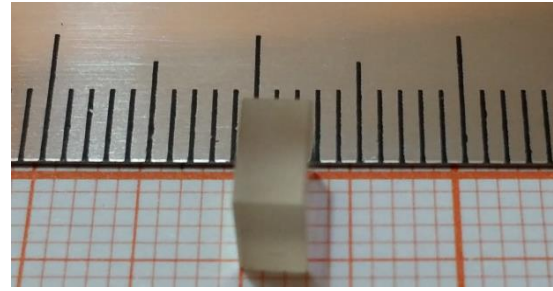
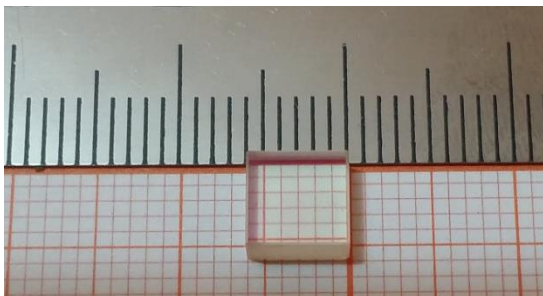


Figure (1) shows the KTiOPO₃ (KTP) crystal 6*6*3mm³

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

$$|d_{eff}| = (d24- d15)\sin 2\theta \sin 2\phi - (d15\sin 2\phi + d24\cos 2\phi)\sin \theta \dots(2-38)$$

The angle θ is measured relative to the z-axis and ϕ is measured in the xy-plane from the x-axis[5].

The Table (1)shows linear and nonlinear optical properties of KTiOPO₃ crystal .

Table 1: linear and nonlinear optical properties of KTiOPO₃ crystal

Linear and Nonlinear Optical Properties	
Thermal Expansion Coefficient	$\alpha_x = 11 \times 10^{-6}/k$, $\alpha_y = 9 \times 10^{-6}/k$, $\alpha_z = 0.6 \times 10^{-6}/k$
Mohs Hardness	5
Density	3.01 g/cm ³
Transparency Range	350~4500nm
Therm-optic Coefficients (/°C)	$dn_x/dT = 6.1 \times 10^{-6}$, $dn_y/dT = 8.3 \times 10^{-6}$ $dn_z/dT = 14.5 \times 10^{-6}$
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: $24^{\circ}\text{C}\cdot\text{cm}^{-1}$ Spectral Acceptance: $0.56\text{ nm}\cdot\text{cm}^{-1}$ Angular Acceptance: $14.2\text{ mrad}\cdot\text{cm}^{-1}$ $(\varphi); 55.3\text{ mrad}\cdot\text{cm}^{-1} (\theta)$ Walk-off Angle: 0.55°
Non-vanished NLO susceptibilities [pm/V]	$d_{31}=6.5\text{ pm/V}$ $d_{24}=7.6\text{ pm/V}$ $d_{32}=5\text{ pm/V}$ $d_{15}=6.1\text{ pm/V}$ $d_{33}=13.7\text{ pm/V}$
Sellmeier Equations (λ in μm)	$n_x^2=3.0065+0.03901/(\lambda^2-0.04251)-0.01327\lambda^2$ $n_y^2=3.0333+0.04154/(\lambda^2-0.04547)-0.01408\lambda^2$ $n_z^2=3.3134+0.05694/(\lambda^2-0.05658)-0.01682\lambda^2$

Second Harmonic Generation

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 (1a) below [N. Bloembergen,2005].

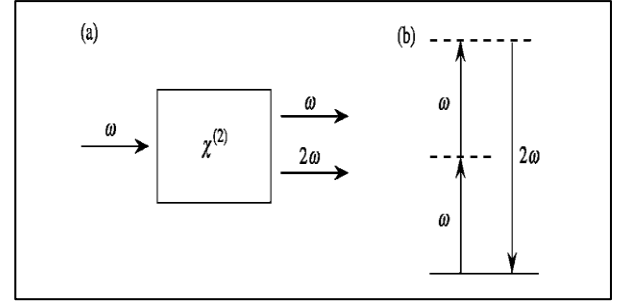


Figure (2) (a) Geometry, of second –harmonic generation. (b) Energy –level diagram, describing, second –harmonic generation[A. Fragemann,2005].

Figure (1-b) 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 [A. Fragemann,2005].

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) [S .Wang,2005]:

$$I = \frac{p}{A} \quad \dots (1)$$

$$A = \frac{\pi w_0^2}{2} \quad \dots (2)$$

Where w_0 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

$$I_{SHG} = d_{eff} L I^2 \sin^2 \left(\frac{\Delta k L_c}{2} \right) \quad \dots (3)$$

When Δk the refractive indices of incident fundamental and second harmonic waves are equal and the intensity of SHG process is maximal.

Therefore, the SHG efficiency can be defined as:

$$\eta = \frac{P_{2\omega}}{P_{\omega}} = \frac{I_{SHG}}{I_s} = d_{eff} L I_0 \sin^2\left(\frac{\Delta k L}{2}\right) \dots (4)$$

Where $P_{2\omega}$ is the SH output power and P_{ω} is the incident beam power [S. Wang, 2005].

Experimental part

The UV-VIS absorption spectra of the KTP crystal was obtained using Mega -2100 Double Beam UV-Vis Spectrophotometer). The nonlinear optical properties for Potassium Titanyl Phosphate (KTiOPO₃ 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). In figure (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 (6x6x3mm³) 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.

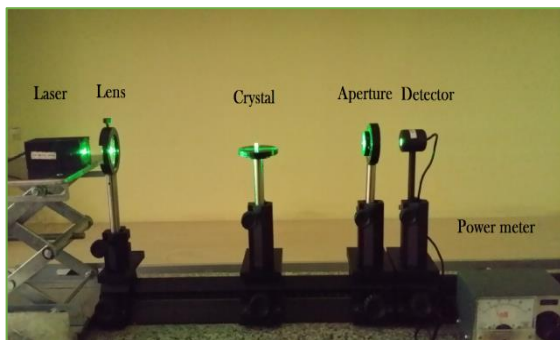


Figure (3) the Z-scan technique set –up experimental in 532nm

In figure (4) shows the experiment of Second Harmonic Generation for KTiOPO₃. 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 (5) 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 (4) the set-up of SHG measurements at wavelength 1064nm



Figure (5) crystal thickness- tilts using bevel protractor.

Result of work and Discussion

The linear absorption spectra of KTP crystal is shown in figure (6). the highest value for the absorbance is 0.312 at wavelength 330 nm. After that, rapidly it decreased with the increased of the wavelength.

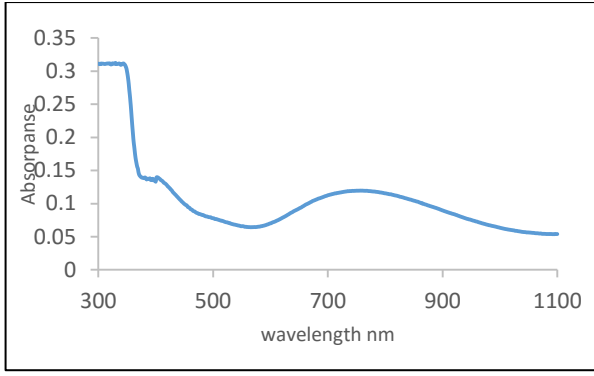
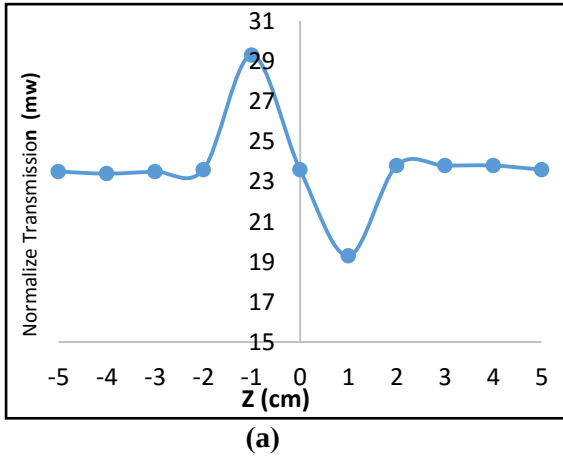
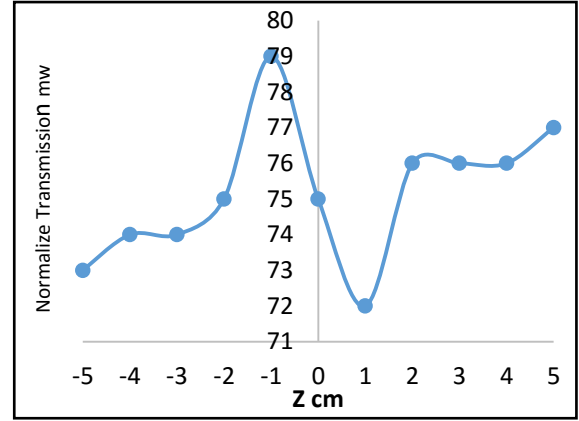


Figure (6): Linear absorption spectra for KTiOPO_3

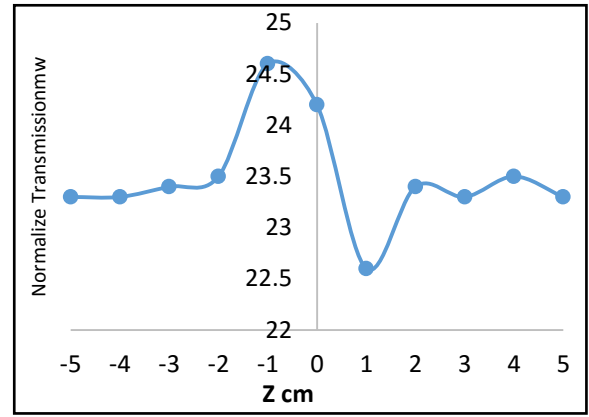
The closed –aperture and open –aperture Z-scan transmittance curves for KTP crystal are shown in figure 7 (a, b, c))and 8 (a, b, c), respectively. the peak followed by valley-normalized transmittance obtained from the close aperture z- scan measurement, indicates that the sign of the refractive index is negative (self-defocusing) for the KTP crystal.



(a)



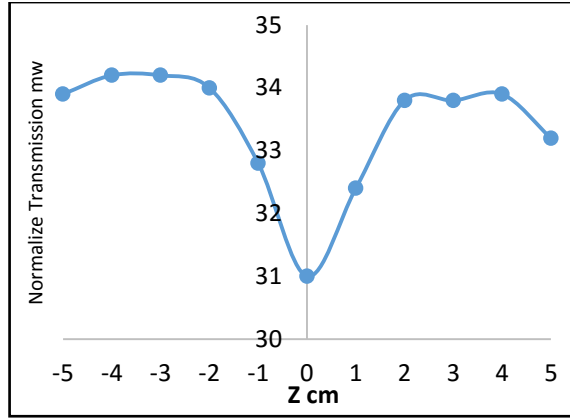
(b)



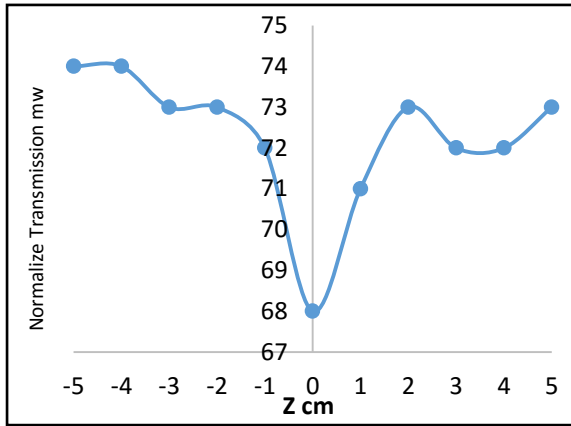
(c)

Figure (7 a, b, c) The normalize transmittance curve as a function of position for the crystal when the close aperture.(a) at wavelength 1064nm with power 35mW.(b) at wavelength 532nm with power 80 mW.(c) at wavelength 532nm with power 25mW.

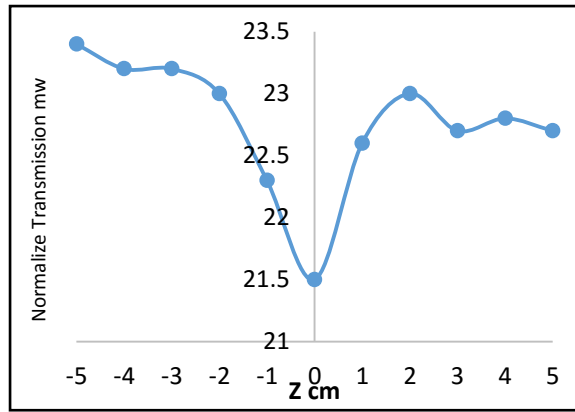
The behavior of open aperture Z-scan transmittance curve in figure 6(a, b, c) indicates that KTiOPO_3 exhibits nonlinear two photon absorption process.



(a)



(b)



(c)

Figure (8 a, b, c) The normalize transmittance curve as a function of position for the crystal when the open aperture.(a) at wavelength 1064nm with power 35mW.(b) at wavelength 532nm with power 80 mW.(c) at wavelength 532nm with power 25mW.

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 $\Delta\Phi_o$,

The relation is define as [D. J.Hagan,2001].

$$\Delta T_{p-v} = 0.406 |\Delta\Phi_o| \quad \dots (1)$$

Where 0.406 constant quantity. And

$$\Delta\Phi_o = k n_2 I_o L_{eff} \quad \dots (2)$$

$\Delta\Phi_o$: nonlinear phase shift, k is wave number [E. Shahriari,2010],

$$k = 2\pi / \lambda \quad \dots (3)$$

The nonlinear refraction index n_2 is given by:

$$n_2 = \Delta\Phi / k I_o L_{eff} \quad \dots (5)$$

I_o is the initial intensity of the laser beam at focus $z=0$ [S. K.saadi, et al,2017].

$$I_o = 2P / \pi w_o^2 \quad \dots (4)$$

Where w_o^2 the radius of laser beam, P is the power of laser beam.

Where L_{eff} is the effective thickness of the sample [S. K.saadi, et al,2017].

$$L_{eff} = (1 - \exp^{-\alpha t}) / \alpha \quad \dots (6)$$

α is linear absorption coefficient, which can be found from the curved Absorption and t is the thickness sample[A. N. Alias et al,2013].

$$\alpha = 2.303 \times A / t \quad \dots (7)$$

A: absorption

The change of the nonlinear refraction index Δn with the laser beam intensity I_o in the following relation [M. Sheik- Bahae,et al,1989]:

$$\Delta n = n_2 I_o \quad \dots (2-30)$$

From the open aperture curve, the nonlinear absorption coefficient β can be calculate from the formula [M. Sheik- Bahae, et al,1990]:

$$\beta = \frac{2\sqrt{2}}{I_0 L_{eff}} \Delta T \quad \dots (2-31)$$

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

Table 2 : summarized the nonlinear optical properties that calculated for KTiOPO3 crystal.

λ (nm)	Power of laser(mW)	$\Delta\Phi$ (Rad)	ΔT_{p-v}	$n_2 \times 10^{-14}$ (cm ² /mW)	$\Delta n \times 10^{-9}$	T_{min}	$B \times 10^{-3}$ (cm/mW)
1064	35	24.137	9.8	3.59	4.08	31.2	2.71
532	80	17.241	7	3.67	5.77	68	4.32
	25	4.926	2	3.35	1.65	21.5	4.37

The figure (3) shows the experimentally results. The intensity of harmonic generation as a function for 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.

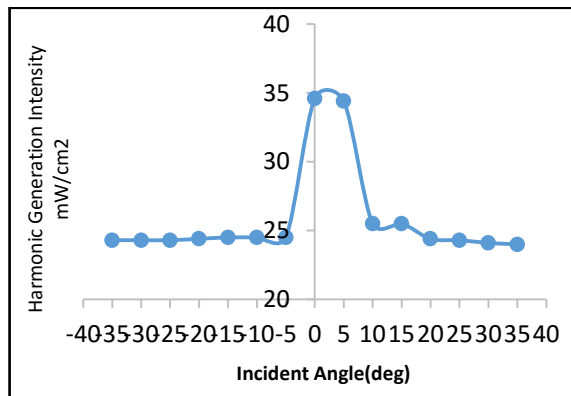


Figure (3) Harmonic generation output intensity versus incident angle

The different values of harmonic generation of intensity at each the input power due to the change 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. 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 0°.

The efficiency of harmonic generation can be calculated from equation (2-16) and (2-38). It can be noticed from the results in Table (3).

Table 3: 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

The result of the real $\text{Re}(\chi^{(3)})$ and imaginary $\text{Im}(\chi^{(3)})$ parts of the third order nonlinear optical susceptibility $\chi(3)$, were

expressed through nonlinear refraction index and nonlinear absorption coefficient, by the following relation [S. A. Aakif,2009]:

$$\text{Re} [\chi^{(3)}] (\text{esu}) = 10^{-4} \frac{\epsilon_0 c^2 n_0^2 n_2}{\pi} (\text{cm}^2/\text{W}) \dots (5)$$

$$\text{Im} [\chi^{(3)}] (\text{esu}) = 10^{-2} \frac{\epsilon_0 c^2 n_0^2 \lambda \beta}{4\pi^2} (\text{cm}^2/\text{W}) \dots (6)$$

Where c: is the speed of the light, n_0 : the linear refractive index, λ : is the wavelength, n_2 : is the nonlinear refractive index, β : is the nonlinear absorption coefficient.

The absolute value of the third order nonlinear optical susceptibility $|\chi^{(3)}|$ can be expressed by the following relation [M. Sheik-Bahae, et al,1989] :-

$$|\chi^{(3)}| = [\text{Re} (\chi^{(3)})^2 + \text{Im} (\chi^{(3)})^2]^{1/2} \dots (7)$$

Table 4: The results of the real and imaginary parts of the third order nonlinear optical susceptibility $\chi^{(3)}$ for KTiOPO₃

λ (nm)	Power of laser (mW)	Re. $\chi^{(3)}$ $\times 10^{-11}$ (esu)	Im. $\chi^{(3)}$ $\times 10^{-3}$ (esu)	$ \chi^{(3)} $ $\times$ 10^{-3} (esu)
1064	35	2.54	2.55	2.55
532	80	2.92	2.28	2.28
	25	2.66	2.32	2.32

Optical limiter is a nonlinear optical process in which the transmittance of a material decreases, when increased incident light intensity [G. S. He,2015].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 [R. A.Ganeev,2013]. One of the major potential applications of these devices is sensor and eye protection. [D. Marciu,1990]

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.

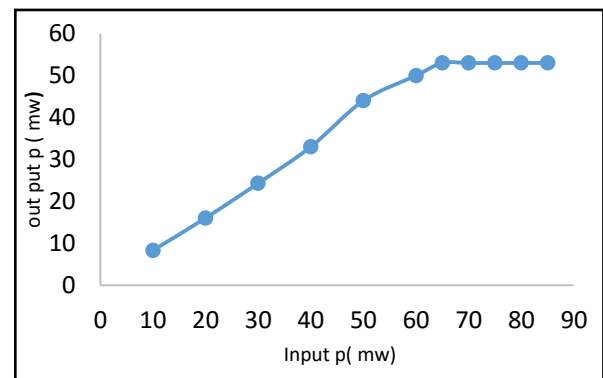


Figure (4-15) The Optical Limiting for KTP crystal.

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. The values optical limiter for KTiOPO₃ crystal listed in table (4).

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

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

Conclusion

The results that we obtained from this study, it is shown that this crystal possesses a high linear properties and a large response to the nonlinear effects, which enables this crystal to be fit material at the nonlinear optical , second harmonic generation applications and optical power limiter.

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