Increasing the conversion efficiency of KTP crystal using the pump-power technique

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Abstract
In this study, the effect of pumping power on the conversion efficiency of nonlinear crystal (KTP) was investigated using laser pump-power technique. The results showed that the higher the pumping power values, the greater the conversion efficiency (η) and, as the crystal thickness increases within limitations, the energy conversion efficiency increases at delay time of (0.333 ns) and at room temperature. Efficiency of 80 % at length of KTP crystal (L = 1.75 X 10^{-3} m) and P_{in} = 28 MW, and also, compare the experimental results with numerical results by using MATLAB program. The contribution of this technique is measuring ultrafast phenomena inside matter such as the movement of atoms or electron excitations.

Key words
Pump-power technique, second harmonic generation, KTP crystal.

Introduction
The pump-p technique are the most ultrafast spectroscopy measurements has brought a better understanding of dynamic processes in many branches of physics, chemistry, and biology. It has been used to discover and monitor fast temporary chemical processes in living cells [1] or in solution [2], also to study fast chemical substance reactions [3-5].

A prevalent type of spectroscopy includes pumping a sample with one light beam and probing the effect of the result on the sample with a second beam, observing, for example, with a beam of probe the variation in absorption or scattering derived by a beam of pump [6]. The experimental methods used in the laser probe technique are determined by determining the time required for an experiment. The "slow" measurements that are determined are the study of the range of interactions with time of approximately <10 ns. Pump sources are controlled independently by using 'real-time' measurements. In short...
periods of time, restrictions are increasing due to inaccuracies in the bombardment of the light source, and the response time of detectors and electronics, to output data. For these reasons, the measurements tend to have a time of <10 ns, which leads to the use of a single light source which works as a pump and probe [7]. The pump-power laser technology is one of the basic techniques used to study all spectra, which has made progress in many scientific fields, and calculate the appropriate parameters for phase matching of biaxial crystals [8]. KTP crystals are used to study nonlinear optical efficiencies [9-11], and the efficiency of SHG in KTP crystallization [12-15].

In this paper, experimentally and numerically the effect of increasing the pumping power ($P_{\text{in}}$) of pulsed laser light on the conversion efficiency ($\eta$) of nonlinear KTP crystal was studied using three different lengths of nonlinear crystal ($L$) at room temperature (RT). Because of the optical properties of KTP crystal is depending on temperature so the conversion efficiency ($\eta$) is depending on it [15]. Moreover, the conversion efficiency ($\eta$) versus cross section area of laser spot ($A$), at length of nonlinear crystal $L=1.75\times10^{-3}$ m, $P_{\text{in}}=3\times10^{7}$ W was studied also. Finally, conversion efficiency ($\eta$) versus mismatching $\Delta k$ at different length of nonlinear crystal $L=1.65\times10^{-3}$ m, $L=1.7\times10^{-3}$ m, and $L=1.75\times10^{-3}$ m, was investigated.

**Theoretical**

Using the wave equation when light is interaction with nonlinear materials, so the generation of radiation with the second harmonic frequency and according to the equation below [12]:

$$\nabla^2 E - \frac{n^2}{c^2} \frac{\partial^2 E}{\partial t^2} = -\frac{4\pi}{c^2} \frac{\partial^2 P_{\text{NL}}}{\partial t^2}$$

(1)

where $n$ and $c$ are the refractive index of the material and the velocity of light in the vacuum, $P_{\text{NL}}$ the polarization associated with the nonlinear response drives the electric field $E$. The equation above gives a fact when $\frac{\partial^2 P_{\text{NL}}}{\partial t^2}$ is non-zero, it accelerates several times and generate accelerated loads of electromagnetic radiation. The second harmonic generation process may be effective, where the incident beam light with frequency $\omega$ on the crystal will be converted to a double-frequency $2\omega$ beam when appropriate conditions are available. The second harmonic generation is the degenerate case of three wave interaction where two of the waves have equal frequencies $\omega_1 = \omega_2 = \omega$ and $\omega_3 = 2\omega$. Therefore, $E_1$ and $E_2$ are the electric fields of the same fundamental beam. Since $E_1 = E_2 = E_{\omega}$ and $E_3 = E_{2\omega}$, two coupled differential equations are [16]:

$$\frac{dE_{\omega}}{dz} = i\omega \sqrt{\frac{\mu_0}{\varepsilon_{\omega}}} d_{ijk} E_{2\omega} E_{\omega}^* \exp(-i\Delta k z)$$

(2)

$$\frac{dE_{2\omega}}{dz} = -2i\omega(1/2) \sqrt{\frac{\mu_0}{\varepsilon_{2\omega}}} d_{ijk} E_{2\omega}^2 \exp(i\Delta k z)$$

(3)

where $\mu_0$ is the permeability of free space, $\varepsilon_{\omega}$ is the absolute frequency of incident beam light, $\varepsilon_{2\omega}$ is absolute frequency of double-frequency and $d_{ijk}$ is the second order polarization tensor. The factor of (1/2) has to be included in Eq.(3) to account for the degeneracy $\omega_1 = \omega_2$. For efficient energy transfer it is necessary that the interacting waves remain in phase. With $k_3 = k_{2\omega}$, $k_1 = k_2 = k_{\omega}$, and $\Delta k = 0$ in $\Delta k = 2k_\omega - k_{2\omega}$ obtained [16]:

$$k_{2\omega} = k_{\omega} + k_{\omega}$$

(4)

Since $k_{\omega} = 2\pi n_{\omega}/\lambda_0$ and $k_{2\omega} = 4\pi n_{2\omega}/\lambda_0$ from the above condition Eq.(4) follows $n_{\omega} = n_{2\omega}$(that’s mean the incident wave and SHG wave must be have the same phase). Therefore, the phase mismatch expressed by $\Delta k$ can
be written in the case of frequency doubling as follows [16]:
\[
\Delta k = \frac{4\pi}{\lambda_0} (n_\omega - n_{2\omega})
\]  
(5)

where \(\lambda_0\) is the wavelength at the fundamental wave in vacuum. In this case can make the assumption that the fundamental beam is not depleted, and therefore \(\frac{dE_{\omega}}{dx} = 0\) in Eq.(2) and only Eq.(3) has to be considered. By observing the tensor properties of \(d_{ijk}\), integration of Eq.(3) yields [16]:
\[
\frac{i_2(L)}{i_1(0)} = \frac{2\Phi_2(L)}{\Phi_1(0)} = \frac{1}{\Delta k} g^2 L^2 \phi_1(0) \sin c^2 \frac{\Delta k L}{2\pi}
\]  
(6)

where: \(g^2 = 4\hbar \omega^3 \eta^2 d^2\), \(\eta = \frac{\eta_0}{n}\),
\(d = \epsilon_0 \text{eff} \), \(\eta_0 = (\mu_0 \epsilon_0)^{1/2} = 377\Omega\),
\(\phi_1 = \frac{\text{photon}}{s}\), \(\phi_1 = \frac{l}{\hbar \omega}\), \(l = \frac{P}{A} \omega\) is the basic frequency, \(d\) is the second order nonlinear coefficient \(10^{-24} - 10^{-21}\) (A.S/V^2), \(\eta\) is the impedance of the medium, \(\eta_0\) is impedance of free space, \(s\) is photon flux, \(P\) is the incident optical power and \(A\) is the cross sectional area. It has large nonlinear optical coefficient, wide angular bandwidth and small walk-off angle. As well as, it has broad temperature and spectral bandwidth. When \(\Delta k\) is constant, the oscillation of the power of the second harmonic is the function of the crystal length. The distance in which the beam maintain coherent is called coherent length. It is measured from inner face of crystal till the position of greatest power of second harmonic as given in following relation [17]:
\[
L_c = \frac{\pi}{\Delta k}
\]  
(7)

So the Eq.(6) becomes:
\[
\frac{i_2(L)}{i_1(0)} = \frac{2\Phi_2(L)}{\Phi_1(0)} = \frac{1}{2} g^2 L^2 \phi_1(0) \sin c^2 \frac{L}{2L_c}
\]  
(8)

Modeling part
Numerically, the MATLAB program was used to calculate conversion efficiency (\(\eta\)) as a function of pumping power (\(P_{in}\)) of pulsed laser light with minimum delay time (0.333 ns) and three different lengths of nonlinear crystal length (\(L=1.65\times10^{-3}, 1.7\times10^{-3}\), and \(1.75\times10^{-3}\)) m at RT, as shown in Fig.1. Table 1 represents the most important elements and parameters that used to investigate the second harmonic generation of KTP crystal using pump-power technique. The value of (L) in step 5 is changed to study the effect of crystal thickness on the calculated crystal efficiency of 15. The selection rules for crystal dimensions are shown according to Eq.(7).

![Fig.1: Program of conversion efficiency of KTP.](image)
Table 1: Some properties of Nd: YAG laser and KTP crystal.

| Element | Properties |
|---------|------------|
| Nd:YAG Laser (Nd:Y3Al5O12) | Fundamental wavelength $\lambda=(1064 \pm 10^2 \text{ m})$ | Half Wavelength (SHG) $\lambda(2\omega)=1064/2 \text{ m}$ |
| | Divergence angle $0.625 \text{ mrad}$ | Linewidth $0.6 \text{ mm}$ |
| | Pulse duration $\tau=10 \times 10^{-9} \text{ s}$ | Maximum Pulse energy $E=1000 \text{ J}$ |
| | Refractive index 1.92 | Reflective index $\rho=4.26 \text{ mm/cm}^3$ |

| KTP crystal (KT/OPO$_4$) | Refractive index of fundamental wavelength $n=1.8597$ | Refractive index of second harmonic wavelength $n(2\omega)=1.8887$ |
| | Phase matching $\theta=90^\circ$ | Phase matching $\phi=23.5^\circ$ |

| | Dimensions $(3 \times 3 \times 1.75 \text{ mm})$ | Nonlinear coefficients $d_{31}=3.64 \times 10^{-12} \text{ (m/V)}$ |
| | | $d_{31}=1.91 \times 10^{-12} \text{ (m/V)}$ |
| | Effective SHG coefficient $\Delta n=-3.30 \times 10^{-13} \text{ (m/V)}$ | |

| | Dimension Tolerance $W \times 1 \times (-0.5 \pm 0.1 \text{ mm})$ | Angle Tolerance $\Delta \theta \leq 0.25^\circ$, $\Delta \phi \leq 0.25^\circ$ |
| | Parallelism $\leq 20 \text{ arc seconds}$ | Perpendicularity $\leq 5 \text{ arc seconds}$ |

- Optical bench
  - Genetic company
  - Dimension $= Length \times Width$
  - $D=59 \times 59 \text{ cm}^2$
- Beam splitter
  - Genetic company
  - 50% $\times$ 50%
- Mirrors
  - Genetic company
  - M1, M2, and M3
- Laser power and energy meter
  - Genetic company
  - Model S6001(R2)
  - Power detection range at 1064$\mu$m $10 \text{ mW}$
  - Digital resolution $1 \text{ mW}$
  - Response time $1 \text{ s}$

Experimental work

In order to study the effect of the pumping power ($P_{in}$) on the conversion efficiency ($\eta$) of KTP crystal, pump-powertechnique was used as shown in Figs.2 and 3. Table 2 represents the most important elements and parameters that used to investigate the second harmonic generation of KTP crystal using pump-power technique. The Nd: YAG laser passively Q-Switched was used with a wavelength of 1064 $\mu$m. This technique requires several steps which are as follows: The first step is dividing the laser beam using a beam splitter into two beam parts. After separating the two pulses (pump and probe pulses), these two pulses each have their own path to reach the crystal KTP, three mirrors called M1, M2, and M3 were put in prop path, which are separated by distances as shown Table 2. It explains all state, where the total path of the probe pulse is 126.5 cm (Where, the total path is the sum of the paths that the laser pulse takes during its reflection in mirrors, starting from the beam splitter and ending with the crystal), while the path of the pump pulse is 25 cm (it is the distance from the beam splitter to the crystal directly), so the delay between the pump and probe pulse is controlled by changing the path length difference between the pump and probe pulse, giving a delay described by:

$$dT = \frac{dx}{c} \quad (9)$$

where $c$ and $dx$ are speed of light and optical path difference respectively. The optical mirrors M1, M2, and M3 can be mounted on translation stages to allow easy variation of path length difference (see Table 2). The process of measuring the energy of the probe is done using a power meter after its interaction with the crystal. To study the effect of delay time ($T$) on conversion efficiency ($\eta$), taking four different values as shown in Table 2. One measures the pump induced
change in the transmission or reflection of the probe beam as a function of the time delay, i.e. as a function of the time delay between the arrival of the pump and probe pulses. This measurement contains information on the relaxation of the electrons in the medium. Due to the dependence of the conversion efficiency along the crystal, three different dimensions were taken for the KTP crystal according to the added Eq.(8). The optical bench dimensions that used in system was 59 x 39 cm.

![Fig.2: pump-power laser technique.](image)

![Fig.3: The pump–probe technique.](image)

| Distance of Beam splitter to M1 (cm) | Distance of M1 to M2 (cm) | Distance of M2 to M3 (cm) | Distance of M3 to KTP crystal (cm) | Total distance of probe pulse (cm) | Distance of pump pulse (cm) | Delay time (T) between two pulses (ns) |
|-------------------------------------|--------------------------|--------------------------|-----------------------------------|----------------------------------|--------------------------|--------------------------------------|
| 21                                  | 38.5                     | 46                       | 21                                | 126.5                            | 25                       | 3.524                                |
| 14.5                                | 20                       | 29                       | 21                                | 84.5                             | 25                       | 2.066                                |
| 5.5                                 | 7.5                      | 14                       | 21                                | 48                               | 25                       | 0.798                                |
| 3                                   | 5                        | 6                        | 21                                | 35                               | 25                       | 0.333                                |
Results and discussion

In Fig.4 can note the significant effect of the length of the KTP crystal on its conversion efficiency, where it is increase very significantly by increasing the length of the crystal to reach 80 % at L=1.75×10^{-3} m and P_{in}=28 MW. The numerical results obtained to calculate the conversion efficiency of KTP crystallization were investigating using MATLAB program, part one where these results are based on the variables and constants in Table 3. The numerically results, of conversion efficiency (\eta) as a function of pumping power (P_{in}) of pulsed laser light with minimum delay time (0.333 ns) and three different lengths of nonlinear crystal length (L=1.65×10^{-3},1.7×10^{-3}, and 1.75×10^{-3}) m at RT, as shown in Fig.4. Experimentally, to study the effect of the nonlinear crystal length L on conversion efficiency (\eta), three different crystals were taken, as shown in Fig.5, where it is represent the conversion efficiency (\eta) versus pumping power (P_{in}) at different length of nonlinear crystal L (1.65×10^{-3} m blue line, 1.7×10^{-3} m red line, and 1.75×10^{-3} m green line and delay time was 0.333 ns and at RT. When compare the numerically result in Fig.4 with experimental results in Fig.5, show the numerical results are close to the experimental results. The experimental conversion efficiency (\eta) were 80% at L= 1.75 x 10^{-3} m, \text{P}_{(in)} = 28MW and equal 83 % numerically. This refers to fewer amounts of error percent between the numerical and experimental result. The achieved results showed that the estimated error is about 3 % for numerically results, in fact, these estimated errors are regarded as fewer values according to the related literatures, which tells that this different in conversion efficiency is due to some little loss of laser beam throughout its interacting with the optical elements of the SHG experiment.

![Fig. 4: Modeling resulting of efficiency (\eta) as a function of pumping power (P_{in}) of pulsed laser light at RT.](image-url)
Table 3: Show most parameters of second harmonic generation setup at RT.

| Speed of light C (m/s²) | Mobility μ₀ (N/A) | Permittivity ε₀ (N/V) | Impedance η (Ω) | The length of nonlinear crystal L(m) |
|-------------------------|-------------------|-----------------------|----------------|-----------------------------------|
| 3×10⁹                  | 1.256×10⁶         | 8.854×10⁻¹²           | 377            | (1.75)×10⁻³                       |
| Refractive index of fundamental wavelength n₀ | Refractive index of second harmonic wavelength n(2ω) | Phase matching Φ (degree) | Phase matching θ (degree) | Nonlinear coefficients (m/V) |
| 1.8297                  | 1.8887            | 21.5                  | 90             | d₃₃ =7.6×10⁻¹²                     |
| Effective SHG coefficient dₑₑ (m/V) | Effective SHG coefficient dijk (mN/V²) | Phase mismatching Δk (m⁻¹) | Fundamental wavelength λω (m) | Half Wavelength λ(2ω) (m) |
| -3.3613×10⁻¹²           | -2.9764×10⁻²⁵     | -6.9682×10⁷           | 1064×10⁻⁹      | 532×10⁻⁹                         |
| Pulse energy E (J) | Pulse duration τ (s) | Power=E/τ (W) | Cross-sectional of spot laser light A (m²) | Time Delay T(ns) |
| (0-300)×10⁻³           | 10×10⁻⁹           | (0-300)×10⁵           | 0.0125×10⁻⁶    | 1.006, 0.798, and 0.333          |

Fig.5: Experimental results of conversion efficiency vs. incident power (W) at different length of nonlinear crystal and at RT.

To study the effect of mismatching Δk on conversion efficiency (η) at different length of nonlinear crystal L equal 1.65×10⁻³, 1.7×10⁻³, and 1.75×10⁻³ m, was investigated numerically using MATLAB program, at RT as shown in Fig.6. The effect of phase mismatching Δk is therefore to reduce the conversion efficiency (η) of SHG by factor sinc² (∆kL/(2π)). This factor is unity for Δk =0 and drop as Δk increases, and vanishing when |Δk| = 2πL as shown in Fig.6. For a given L, the mismatch Δk corresponding to a prescribed to L, so that the phase matching requirement becomes more significant as L increase. For a given mismatch Δk, the length Lₖ = (2π|Δk|) is measure of the maximum length with
in which SHG is efficient, $L_c$ is often called the coherence length. This figure exponential decay curve where to maximize the SHG conversion efficiency we must confine the wave of laser spot to the smallest possible area $A$ and the largest interaction length $L$. Moreover, the conversion efficiency ($\eta$) versus cross section area of laser spot ($A$), at length of nonlinear crystal $L=1.75\times10^{-3}$ m and $P_{in}=3\times10^7$ W was studied numerically also at RT, as shown in Fig.7. Finally, the conversion efficiency ($\eta$) versus length of nonlinear crystal with cross section area $0.125\times10^{-8}$ m$^2$ of laser spot, and $P_{in}=3\times10^7$ (W), at RT was studied numerically as shown in Fig.8. Actually, the achieved results ensure the correct path of the research strategy and acceptable results that can be apply this achievement to safely transfer into application. Fig.8 shows the SHG conversion efficiency for KTP in dependence on the crystal length. From this curve the optimum crystal length for SHG conversion was extracted to be $1.8 \times 10^{-3}$ m for cross section area $0.125 \times 10^{-8}$ m$^2$ of laser spot and pumping power $3 \times 10^7$ W at R.T.

![Fig. 6: Factor $\text{sinc}^2(\Delta kL/2\pi)$ Vs. $\Delta k/2\pi$ at different length of nonlinear.](image-url)
Conclusion

In this work we conclude that the power conversion efficiency depends on several factors as it increases either by increasing the laser pumping energy, or by increasing the length of the crystal at a constant temperature and at a constant delay time between two pulses by using laser pump-power technique, or by changing phase matching. The pump-power technique enables us to measure ultrafast phenomena inside matter such as the movement of atoms or electron excitations.

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