International Journal of Science and Research (IJSR)
ISSN (Online): 2319-7064
Index Copernicus Value (2013): 6.14 | Impact Factor (2015): 6.391

Study of the Second and Third Harmonics Generation in Lithium Triborate Single Crystal

Zahraa S. Shanon[1], Raad Sh. Alnayli[2], Khawla J. Tahir[3]

[1, 2] University of Al-Qadisiyah, College of Education, Department of Physics, Al-Qadisiyah, Iraq
[3]University of Karbala, College of Sciences, Department of Physics, Karbala, Iraq

Abstract: ***In this paper, we investigate to approach the ideal conditions to optimized the performance of the single pure Lithium Triborate crystal (LBO) (The boron–oxygen compound LiB_3O_5 is recognized as a new Nonlinear Optical crystal). Which has dimension (5.95×5.95 ×5.95) mm^3 as proposal element model for the combination of the Second Harmonics Generation (SHG) and Third Harmonics Generation with Nd Yag laser has the spectral wave length around 1.064 μm and 532 nm. The main results are very satisfactory apply this proposal model as optimum controlling element for the combined application as, the second harmonics generator (SHG) and the Third Harmonics Generation at 1.064μm and 532 nm.***

Keywords: second-harmonic generation, third-harmonic generation, cubic susceptibility $\chi^{(3)}$,Lithium Triborate

1. Introduction

One of the most important nonlinear optical processes for technical applications is the generation of harmonics from laser light. Presently it is one of the main applications of nonlinear optics, maybe the only really important one. We will explain here second-harmonic generation and third harmonic generation, widely used for producing visible and near ultraviolet coherent light. [1]

The nonlinear optical (NLO) processes in crystals through third-order NLO susceptibility $\chi^{(3)}$ are of great research and practical interest because of their applications to various optical systems, optical information processing and optical functional devices. The generation of optical harmonics is well established spectroscopy method to obtain valuable information on NLO properties of the materials. Interesting results have been reported about third-harmonic generation (THG) in crystals, concerning the determination of their cubic susceptibility $\chi^{(3)}$. The THG method is simple and reproducible because the nonlinearity induced by the thermal effect does not interfere with the THG signal. On the other hand, the spectroscopy of two-photon absorption (TPA) provides also data concerning $\chi^{(3)}$ and the electronic transitions in crystals by two-photon resonances[2]. In this paper, we report the experimental results obtained on the third-order NLO response of lithium triborate crystal. The aim of the present work is to study the second and third-order NLO response of the Lithium Triborate crystal.

2. Materials and Methods

2.1 Lithium Triborate crystal (LBO)

LiB_3O_5 single crystals are orthorhombic with four formula units per unit cell. The space group and the point group are $Pna2_1$ (C_{2v}^9) and mm2, respectively, and the lattice parameters are a=8.4773 Å, b=7.3788 Å, and c=5.1395 Å[47]. The structural unit of lithium triborate (LiB_3O_5) consisted of the anionic group $(B_3O_7)^{5-}$ with one BO_4 tetrahedron and two BO_3 triangles[48], Figure 1 shows a projection of the LiB_3O_5 structure onto the (010) and the(001) planes. The boron atoms are located within coordination polyhedra of two types, triangles [B(1) and B(2)] and tetrahedra [B(3)]. The B(1) and B(2)atoms lie within planar triangles formed by oxygen, and the B(3) Atoms within oxygen tetrahedra. The average nearest neighbor distance of B(1) -O is 1.3692 Å, of B(2)-O is 1.3713 Å, and of B(3) -O is 1.473 Å. There are four Li atoms in a cell, and they are surrounded by four oxygen atoms in a considerably distorted tetrahedron. The Li–O distance is rather longer than the B–O distance and ranges from 1.9887 to 2.1722 Å with a distorted tetrahedral coordination [3].

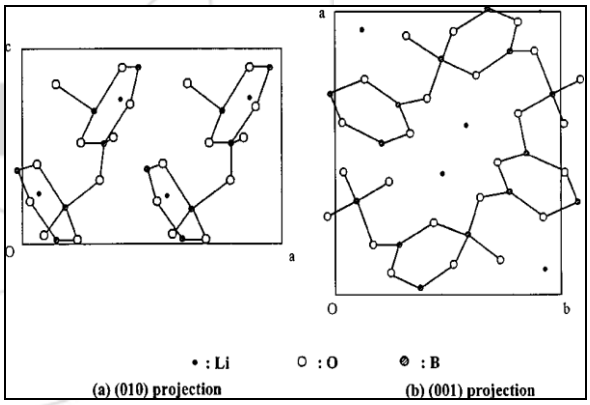

Figure 1: Projection of the crystal structure of a LiB_3O_5 single crystal.[4]

2.1.1 The transmission property of LBO

LBO crystal have wide range of transparency between (0.16 - 2.6μm) as see in figure 2. this feature of LBO is ahead of other borate crystals for large transparency at the practical wavelength, LBO crystal having much higher pulse energy and high average power of the Nd:YAG laser because of the Nd:YAG laser damage threshold was about 40 GW/cm^2.[5]

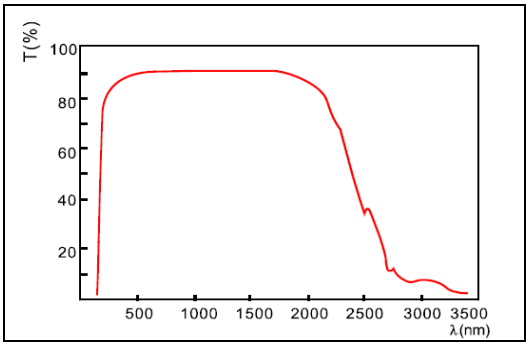

Figure 2: Transparency curve of LBO[6]

2.1.2 Second Harmonic Generation (SHG) and Third Harmonic Generation (THG) at Room Temperature:

For type I and type II of LBO interaction is phase matchable for the Second Harmonic Generation and Third Harmonic Generation of Nd:YAG and Nd:YLF lasers, So the SHG at room temperature, type I phase matching can be satisfy and has the maximum effective SHG coefficient in the principal XY and XZ planes (see Fig. 3) [7], in a wide wavelength range from 551 nm to about 2600 nm (the effective SHG coefficient see Table 1). The best phase matching of type II falls in the principal YZ and XZ planes (see Fig.(3) - the effective SHG coefficient see Table 1). more than 70% SHG conversion efficiencies at pulsed and 30% for CW Nd:YAG laser, and THG conversion efficiency

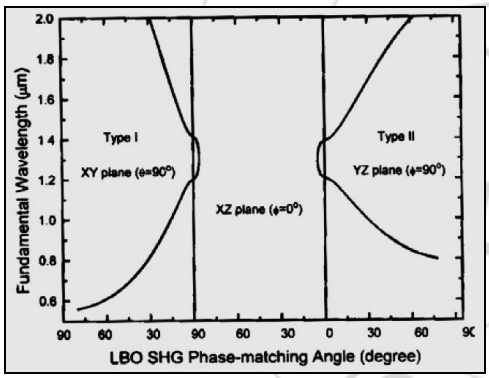

Figure 3: SHG tuning curves of LBO.[7]

Table 1: Optical and Nonlinear Optical Properties[8]:

Linear Optical Properties	
Transparency Range	160-2600nm
Refractive indices: at 1064 nm at 532 nm at 355 nm	 $n_x = 1.5656$, $n_y = 1.5905$, $n_z = 1.6055$ $n_e = 1.5785$, $n_o = 1.6065$, $n_z = 1.6212$ $n_e = 1.5971$, $n_o = 1.6275$, $n_z = 1.6430$
Therm-optic Coefficient (/ °C, λ in μm)	$dn_x/dT=-9.3*10^{-6}$ $dn_y/dT=-13.6*10^{-6}$ $dn_z/dT=(-6.3-2.1\lambda)*10^{-6}$
Sellmeier Equations (λin μm)	$n_x^2=2.454140+0.011249/(\lambda^2-0.011350)-0.014591\lambda^2-6.60x10^{-5}\lambda^4$ $n_y^2=2.539070+0.012711/(\lambda^2-0.012523)-0.018540\lambda^2+2.00x10^{-5}\lambda^4$ $n_z^2=2.586179+0.013099/(\lambda^2-0.011893)-0.017968\lambda2-2.26x10-5\lambda^4$
Nonlinear Optical Properties	
SHG Phase Matchable Range	551-2600nm (Type I) 790-2150 nm (Type II)
Acceptance Angles(@1064nm)	6.54mrad·cm (ϕ, Type I, SHG) 15.27mrad·cm (θ, Type II, SHG)
Temperature Acceptance	4.7°C·cm(Type I, 1064 SHG) 7.5°C·cm(Type II,1064 SHG)
Spectral Acceptance	1.0nm·cm (Type I, 1064 SHG) 1.3nm·cm (Type II,1064 SHG)
Walk-off Angles (@1064 nm)	0.60°(Type I SHG) 0.12°(Type II SHG)
NLO Coefficients0.60	$d_{eff}(I)=d_{32}\cos\varphi$(Type I in XY plane) $d_{eff}(I)=d_{31}\cos^2\theta+d_{32}\sin^2\theta$(Type I in XZ plane) $d_{eff}(II)=d_{31}\cos\theta$(Type II in YZ plane) $d_{eff}(II)=d_{31}\cos^2\theta+d_{32}\sin^2\theta$(Type II in XZ plane)
Non-vanished NLO susceptibilities	$d_{31}=1.05 \pm 0.09$ pm/V $d_{32}= -0.98 \pm 0.09$ pm/V $d_{33}=0.05 \pm 0.006$ pm/V
Electro-optic coefficients	$\gamma_{11} = 2.7$ pm/V, γ_{22}, $\gamma_{31} < 0.1\gamma_{11}$
Conversion Efficiency	>90% (1064 -> 532nm) Type I SHG
Damage threshold at 1064nm at 532nm at 355nm	 45 GW/cm^2 (1 ns); 10 GW/cm^2 (1.3 ns) 26 GW/cm^2 (1 ns); 7 GW/cm^2 (250 ps) 22 GW/cm^2

2.2 Second Harmonic Generation (SHG)

Second Harmonic generation (SHG) was the first experiment in the history of nonlinear optics discovered by Franken *et al.* [9] soon after the invention of the Ruby laser [10]. The process occurs within a nonlinear medium, usually a crystal (KDP-Potassium di hydrogen phosphate, KTP-Potassium Titanyl Phosphate, etc.), Frequency doubling processes are commonly used to produce green light (532nm) using, a Nd:YAG (Neodymium: Yttrium Aluminum Garnet) laser operating at 1064 nm [11].Second harmonic generation (SHG), In otherwise called "frequency doubling", is a process which converts light of one particular frequency to light at exactly double that frequency. Another way of saying it is to say that the wavelength is halved. This process only takes place under very specific conditions within a material which has a "second-order non-linear" characteristic[12]. Materials of this kind are sometimes called chi-squared (χ^2) materials. It can be produced double frequency by double the energy. Thus two photons of a particular energy (and frequency) are combined (through interaction with a material) to give out a third photon containing all the energy of the original two (at double the frequency). Chemical bonds between pairs of atoms (called dipoles) resonate at particular frequencies (wavelengths, energy levels). [9]

The process of second harmonic generation involves the interaction of two waves at frequency ω to produce a wave with the frequency 2ω. It is schematically illustrated in Fig. (4) below. [12]

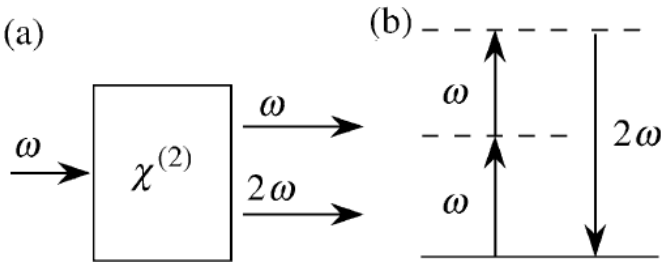

Figure 4: (a) Geometry of second-harmonic generation. (b) Energy-level diagram describing second-harmonic generation.[14]

This Figure explain that the energies of the two absorbed photons don't have to be the same. This is the general case of "three-wave mixing" where two photons combine through interaction with a material to produce a third photon with the sum of the energies of the original photons. However, in the case of SHG we have two photons with the same energy level combining to produce a third with the sum of the energies of the two original photons.[14]

2.3 Third Harmonics Generation (THG):

Third Harmonic Generation THG is the coherent conversion of light with frequency ω into light with frequency 3ω, wavelength λ/3, in material that undergoes intense irradiation. It involves the absorption of three identical photons of energy $\hbar\omega$ and the emission of a single photon of energy $3\hbar\omega$ within the temporal uncertainty interval of $\omega^{-1}\sim10^{-16}$ s. The resultant light propagates in the forward direction. [15] A susceptibility tensors, $\chi^{(n)}$, described the material's susceptibility to a given nonlinear conversion process, this susceptibility relate the polarization field P, induced in the material to the electric field E, of the incident photon.so the third-rank tensor status of the susceptibility tensor to be suppressed and the THG polarization field $P^{(3)}(3\omega)$ to be expressed as:

$$P^{(3)}(3\omega) = \chi^{(3)}(3\omega)\, E^3 \qquad (1)$$

$P^{(3)}(3\omega)$ is the third-order nonlinear polarization.

In a solution or other isotropic media, the measured value for $\chi^{(3)}(3\omega)$ is averaged over orientation and equal to 1/3 $\chi^{(3)}{}_{xxxx}(3\omega)+\chi^{(3)}{}_{yyyy}(3\omega)+\chi^{(3)}{}_{zzzz}(3\omega)$. The susceptibility $\chi^{(3)}(3\omega)$is the term we seek to measure. [18]Third harmonics were generated through two steps with second harmonic generation in a first crystal and sum frequency generation in a second crystal. [16]Third Harmonic Generation (THG) is a nonlinear process, in which incident high intensity laser radiation at the frequency ω, interacting with a nonlinear medium, results in the generation of an additional spectral component at the frequency 3ω. For a tight focused beam it can be generated only on the interface between two media.[17] The process of third harmonic generation produce by a one step in a single crystal. This means that only one optical component is required, which makes for a very simple setup[18]. (see Fig. 5).

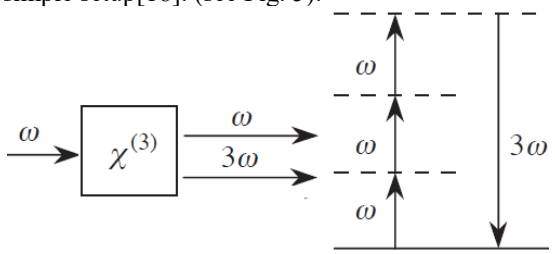

Figure 5: Third-harmonic generation. (a) Geometry of the interaction.(b) Energy-level description. [19]

3. Experimental Setup

3.1 Second and Third Harmonic Generation setup :

Assuming a plane wave and non-depleting fundamental power, the THG output power was measured for different incident angles in the range from -30° to 30° increments of 5°step. The measurements of intensity based on one value of sample thickness 5.99mm. The incident angle of undamental beam θ was changed by tilt the sample forward to the left of the optical axis to make a positive angle and backward to the right of the optical axis to make a negative angle. Bevel-square was used to measure the incident angle as shown in figure (6) and the Figure (7) shows the block diagram of THG measurements.

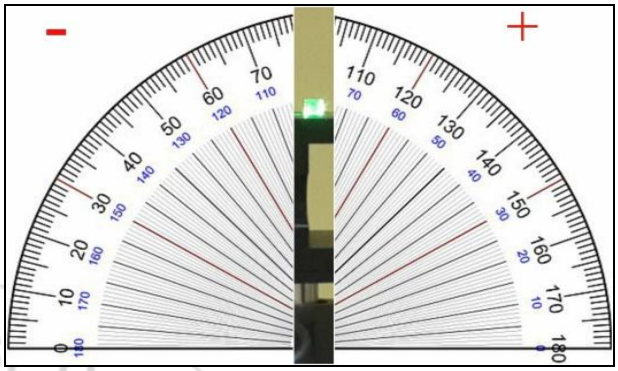

Figure 6: (5.95mm) crystal thickness tilts using bevel-square

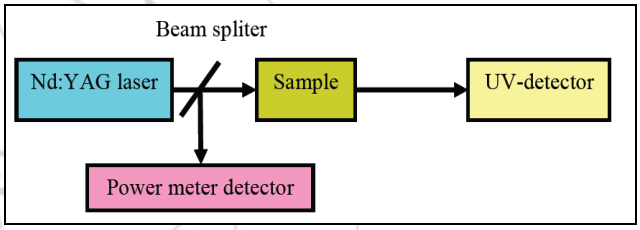

Figure 7: Block-diagram of THG measurements.

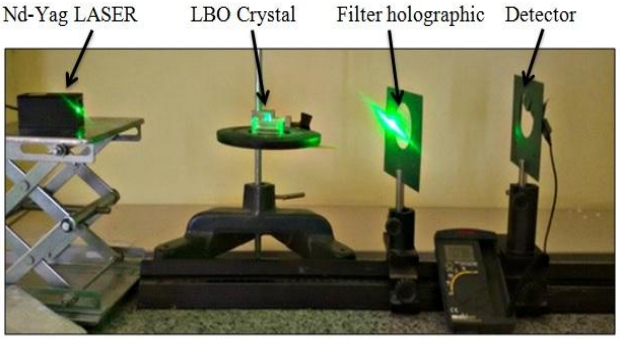

Figure 8: The Set-Up of THG Measurements

4. Result and Conclusion

The Second and Third Harmonic Generation were investigated for Lithium Triborate (LBO) crystal and the thickness of the crystal was (t=5.95mm) with power (P = 25 and 40 mW) at each (532 and 1064 nm) of Nd-YAG laser.

4.1 The Intensity of Third Harmonic Generation (THG):

From the figures (9) and (10), low THG intensity obtained at the angles -20°, 25°. The reason of this effect is that the high re-absorption of THG beams in the LBO crystal at these angles. Furthermore, the path length inside the sample was decreased when increasing the incident angle. THG intensity was vanished at the critical incident angles, which was

obtained at -40° and 40 °because no signal will be detected at the critical angles.

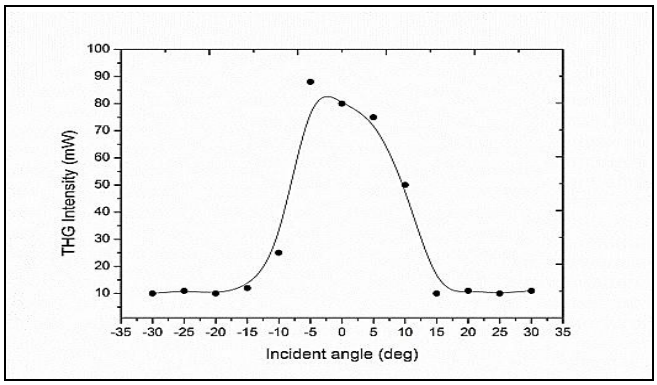

Figure 9: THG output intensity excited at 80mW input power versus incident angle

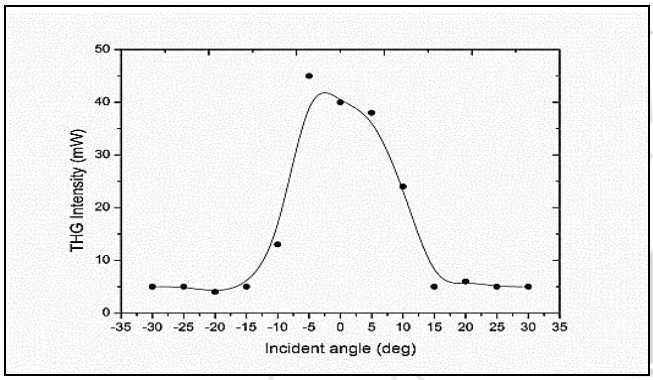

Figure 10: THG output intensity excited at 35 mW input power versus incident angle

The result shows different values of THG intensity at each input power due the change in the incident angle of fundamental beam. The reason of change the incident angle of the fundamental beam is to obtain on efficient THG intensity. The change in the incident angle of fundamental beam was induced change in refractive index. The optimum value of THG intensity was found for each input power and incident angle as shown in table 2:

Table 2: THG intensity excited at 80mW input power incident angle

Input power (mW/cm^2)	Optimum value of $I_{3\omega} \times 10^{11} (mW/cm^2)$	Incident angle θ (degree)
49.1×10^3	1.04	18°
78.6×10^3	7.51	18°

The result of the real and imaginary parts of the third order nonlinear optical susceptibility $\chi^{(3)}$ were determined from experimental determination of n_2 and β according to the following relations[20]:

$$\mathrm{Re}\,\chi^{(3)}(esu) = 10^{-4}\frac{\varepsilon_0 c^2 n_0^2}{\pi}\left(\frac{cm^2}{W}\right) \quad (2)$$

$$\mathrm{Im}\,\chi^{(3)}(esu) = 10^{-2}\frac{\varepsilon_0 c^2 n_0^2 n_2 \lambda \beta}{4\pi^2}\left(\frac{cm^2}{W}\right) \quad (3)$$

The absolute value of was calculated from the following relation :

$$\left|\chi^{(3)}\right| = \left[(Re(\chi^{(3)}))^2 + (Im(\chi^{(3)})^2\right]^{\frac{1}{2}} \quad (4)$$

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

Re. χ^3	Im. χ^3	χ^3
6.75029×10^{-5}	2.97435×10^{-11}	6.75029×10^{-5}
5.74437×10^{-5}	3.72098×10^{-11}	5.74437×10^{-5}

References

[1] D. Kornack and P. Rakic, 2001, "Cell Proliferation without Neurogenesis in Adult Primate Neocortex," Science, vol. 294, Dec., pp. 2127-2130, doi:10.1126/science.1065467.
[2] S. M. Metev and V. P. Veiko, 1998, Laser Assisted Microtechnology, 2nd ed., R. M. Osgood, Jr., Ed. Berlin, Germany: Springer-Verlag.
[3] In Gyoo Kim, Sung Ho Choh and Jung Nam Kim, "NMR Study of ^{7}Li and $^1{}_1$B Nuclei in Nonlinear Optical LiB_3O_5 Single Crystal", Journal of the Korean Physical Society, Vol. 32, pp. S669_S671, (1998).
[4] Ae Ran Lim, Ji Won Kim and Choon Sup Yoon, "Local structure of LiB_3O_5 single crystal from 7Li nuclear magnetic resonance, Journal Of Applied Physics", Volume 94, Number 8,P1, (2003).
[5] Senthilkumar M, 2010, "Crystal Growth and Characterization of $La_2CaB_{10}O_{19}$, $Nd_xLa_{2-X}CaB_{10}O_{19}$and $K_2B_4O_{11}H_8$ for NLO and Laser applications and Theoretical Analysis of Nonlinear Optical Property of $La_2CaB_{10}O_{19}$ Crystal", Thesis, Anna University.
[6] Tolga Depci, "Synthesis and Characterization of Lithium Triborate By Different Synthesis Methods and Their Thermoluminescent Properties", Thesis, Middle East Technical University,2009.
[7] Shen.Y.R,"The principles of nonlinear optics", A Wiley-Interscience Publication John Wiley & Sons, New York, (1984).
[8] Kostyantyn Sukhoy, 2011,"Generation of green second harmonic radiation in LBO, BBO, KTP, and PPLN crystals using passively Q-switched sub-nanosecond microchip laser", Thesis, University of Manitoba.
[9] Luca Rigamonti, Schiff Base, "Metal Complexes For Second Order Nonlinear Optics", Università Di Milano, (2010).
[10] T.H. Maiman, "Stimulated Optical Radiation in Ruby", Nature, 1960, Vol.187, No. 4736, PP 493–494.
[11] Amnon Yariv, Pochi Yeh, "Optical waves in crystals propagation and control of Laser radiation", California,(1983).
[12] Harry J. R. Dutton, "Understanding Optical Communications, International Technical Support Organization", First Edition,1998.
[13] Sergey A. Ponomarenko, "Fundamentals of Nonlinear Optics", Dalhousie university,PP 77, (2015).
[14] Robert W. Body, "Nonlinear Optics", Third Edition, Rochester, New York, 2007.
[15] G. Omar Clay, Andrew C. Millard, Chris B. Schaffer, Juerg Aus-der-Au,Philbert S. Tsai, Jeffrey A. Squier,David Kleinfeld, "Spectroscopy of third-harmonic generation: evidence for resonances in model compounds and ligated hemoglobin", J. Opt. Soc. Am. B,Vol. 23, No. 5, (2006).

[16] Arne Potreck, 2014, "Nonlinear Optical Frequency Conversion for Lasers in Space", thesis, Eberhard-Karls Universität Tübingen, PP 43.