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

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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 LiB₃O₅ is recognized as a new Nonlinear Optical crystal). Which has dimension (5.95×5.95 ×5.95) mm³ 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 χ(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 χ(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 χ(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 χ(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)

LiB₃O₅ single crystals are orthorhombic with four formula units per unit cell. The space group and the point group are Pna2₁ (C₄ᵥ) 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 (LiB₃O₅) consisted of the anionic group (B₂O₅)²⁻ with one BO₃ tetrahedron and two BO₃ triangles[48]. Figure 1 shows a projection of the LiB₃O₅ 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 LiB₃O₅ single crystal.[4]
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

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 ($\chi^2$) 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 $\omega$ to produce a wave with the frequency $2\omega$. It is schematically illustrated in Fig. (4) below. [12]

![Image](image_url)
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 $\omega$ into light with frequency $3\omega$, wavelength $\lambda/3$, 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 $\omega^{-1} \sim 10^{-16}$ s. The resultant light propagates in the forward direction. [15] A susceptibility tensors, $\chi^{(3)}$, describe 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:

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

$P(3)(3\omega)$ 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 $1/3 \chi^{(3)}_{xxx}(3\omega)+\chi^{(3)}_{yyy}(3\omega)+\chi^{(3)}_{zzz}(3\omega)$. 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 $\omega$, interacting with a nonlinear medium, results in the generation of an additional spectral component at the frequency $3\omega$. 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).

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 fundamental beam $\theta$ 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](5.95mm) crystal thickness tilts using bevel-square

![Figure 7: Block-diagram of THG measurements.](5.95mm) crystal thickness tilts using bevel-square

![Figure 8: The Set-Up of THG Measurements](5.95mm) crystal thickness tilts using bevel-square

4. Result and Conclusion

The Second and Third Harmonic Generation were investigated for Lithium Triborate (LBO) crystal and the thickness of the crystal was $(t=5.95\text{mm})$ with power $(P = 25$ and $40 \text{ mW})$ at each $(532$ and $1064 \text{ nm})$ 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 an 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>
<thead>
<tr>
<th>Input power (mW/cm²)</th>
<th>Optimum value of I₃ω (mW/cm²)</th>
<th>Incident angle θ (degree)</th>
</tr>
</thead>
<tbody>
<tr>
<td>49.1 × 10⁴</td>
<td>1.04</td>
<td>18°</td>
</tr>
<tr>
<td>78.6×10⁴</td>
<td>7.51</td>
<td>18°</td>
</tr>
</tbody>
</table>

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

The result of the real and imaginary parts of the third order nonlinear optical susceptibility χ(3) were determined from experimental determination of n² and β according to the following relations [20]:

\[ \text{Re} \chi^{(3)}(esu) = 10^{-4} \frac{2eC^2\beta \lambda^3}{\lambda B} \text{ (cm}^2\text{)} \] (2)

\[ \text{Im} \chi^{(3)}(esu) = 10^{-5} \frac{eC^2\beta \lambda^3}{\lambda B} \text{ (cm}^2\text{)} \] (3)

The absolute value of was calculated from the following relation:

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

Table 3: The result of the real and imaginary parts of the third order nonlinear optical susceptibility χ(3)

<table>
<thead>
<tr>
<th>Re. χ(3)</th>
<th>Im. χ(3)</th>
<th>χ(3)</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.75029×10⁻⁴</td>
<td>2.97435×10⁻¹</td>
<td>6.75029×10⁻⁴</td>
</tr>
<tr>
<td>5.74437×10⁻²</td>
<td>3.72098×10⁻¹</td>
<td>5.74437×10⁻²</td>
</tr>
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</table>

References


