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Second Harmonic Generation of 266 nm Laser Beam Using BBO Nonlinear Crystal

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Abstract

Second harmonic generation (SHG) is a phenomenon observed in nonlinear optics that leads to frequency duplication for a high intensity laser incident on nonlinear crystal using BBO crystal. The SHG yield is achieved when the photons interact with a nonlinear optical material and effectively combine to form new photons with double frequency, and therefore double energy and half wavelength. This paper is concerned with the establishment of an SHG experiment to govern the process of producing half-wavelength laser beam from the input one. The theoretical effort was extended to compute the efficiency by using MATLAB software based on mathematical relationships. The values of the conversion maximum efficiencies, which were computed as a function of the input and output powers of the theoretical computations, were 15.6% and 16% at input and output power values of 0.6 and 0.1, respectively. The experimental results of the laser source of 532nm wavelength (fundamental frequency was 0.563x1015s-1) gave a half wavelength of 266nm (double frequency was 1.126x1015s-1). The conversion efficiencies, computed as a function of the input and output powers of the experimental measurements, were 14.32% and 12.97%, respectively.

Keywords: second harmonic generation, nonlinear optics, nonlinear crystal material

التوليد التوافقي الثاني من شعاع الليزر 266 نانومتر باستخدام بلورة BBO غير الخطية

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الخلاصه

ان التوليد التوافقي الثاني (SHG) هو ظاهرة موجودة في البصريات غير الخطية التي تؤدي إلى تكرار التردد الليزر عالي الكثافة على البلورة غير الخطية باستخدام بلورة BBO. ينمو SHG عندما تتفاعل الفوتونات مع مادة بصرية غير خطية وتتحد بشكل فعال لتشكيل فوتونات جديدة بتردد مزدوج ، وبالتالي مضاعفة الطاقة ونصف الطول الموجي. يهتم هذا البحث بإنشاء تجرية SHG للتحكم في عملية إنتاج شعاع ليزر بطول نصف موجة من المدخلات. لتقدير النتائج الصحيحة. يتم توسيع الجهد النظري لحساب الكفاءة باستخدام برنامج MATLAB على أساس العلاقات الرياضية. كانت الكفاءات القصوى للتحويل المحسوبة كدالة لقدرات المدخلات والمخرجات للحسابات النظرية 15.6 ٪ و 16 ٪ عند 0.0 و 0.1 طاقة المدخلات والمخرجات على التوالي. أعطت النتائج المصدر الليزر بطول موجة (532) mnالتردد الأساسي (

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1–1.563x1015sفصف طول موجة 266) mmالتردد المزدوج .(1–1.126x1015s كانت كفاءات التحويل المحسوبة كدالة لقوى المدخلات والمخرجات للقياسات التجريبية 14.32٪ و 12.97٪ على التوالي

1- Introduction

Nonlinear optics is an important field in physics due to a wide range of recent applications that are related to the propagation of electromagnetic waves. It is based on studying the interaction of high intensity light with matter during its propagation [1]. One of the most important physical phenomena in the nonlinear optics is the second harmonic (SH), which is a useful tool to change the wavelength of the laser beam from one value to another. A high-intensity light source is needed to generate SH, which can be achieved only by using lasers [2]. Nonlinear optics is practically employed to change the wavelength of the laser to achieve increased advantages of these rays. Through this technique, wavelengths longer or shorter than that of the laser pump can be obtained. This technique leads to save the time, effort, and cost in manufacturing laser devices of desired wavelengths. Hence, the second harmonic generation (SHG) became an important part of nonlinear optics which opened new fields and continuous researches to obtain the highest conversion rate and maximum possible energy at the new required wavelength of coherent light [3]. Ossi et al., in 2008, reported a built-in short pulse UVlaser of 1.9ns at 236nm, in which a quasi-three-level of passive Q-switching of 946nm Nd-YAG laser is acting as a pumping source. UV laser pulses were made by two successive events of single-pass frequency duplication. The first is extended along the range of IR up to blue using BIBO crystal, while the second is working along the range of blue up to DUV using β -BBO crystal. The efficiency of conversion for 946nm to 473nm was about 28%, whereas it was 7% for blue to DUV. The average power of UV was about 7.6mW with 230nJ energy and 120W peak power [4]. Fiebig, in 2009, demonstrated a continuous-wave of 490nm wavelength and 1W energy that was set up on 2.5cm3 micro-optical bench using SHG of a single path with a periodic poled bulk crystal of type MgO:LiNbO3. This experiment resulted in a 1W blue light with an optical conversion efficiency of 11%. The module showed an output power stability of higher than 2% [5]. Wang et al., in 2017, reported the structure of a high power blue laser beam of a 250W (i.e., 445nm) in processing materials. The rate of absorption of this laser system supplied on steel was 2.75 times that of the system of single-mode fiber laser operated at 1070nm. The steel characteristics after laser irradiation are commonly estimated to validate the potential of the used high-power blue laser to process materials such as cladding and heat treatment [6]. Xuan et al., in 2018, showed the generation of the deep ultraviolet lasers using solid-state/fiber lasers to process the nonlinear fundamental frequency conversion. The deep ultraviolet laser of 258nm provided low average power of beam quality of about $M_{2<1.5}$ by fourth harmonic generation (FHG). Also, the average power at 193nm was about 1W, which was achieved by SHG. Experiment principles of diamond Raman laser was proved at 23% conversion efficiency by pumping second stokes wavelength, which refers to the great potential of the 193nm DUV laser of generating higher power [7].

2-Materials and methods

2.1 Second Harmonic Generation

The proposed SHG model is based on the laser-material interaction that generates SHG in nonlinear crystal. This depends on introducing an interaction between laser beam and nonlinear crystal which leads to an expected reaction impact of the wavelength of the incident laser. When the laser beam incident occurs on the nonlinear crystal, the frequency of the laser beam is duplicated, which implies obtaining a new shorter laser wavelength, and so on one can choose the laser source and proper nonlinear crystal to achieve the intended short wavelength laser. In SHG, the incident laser generates a nonlinear polarized beams that are oscillating with double the fundamental frequency. Maxwell's equations state the nonlinear polarized waves that are radiating electromagnetic field of twice the frequency. The phase-matching renders the second-harmonic generated field dominantly propagating in the direction of the nonlinear polarization wave [8].

2.2 Theoretical Concepts

The concepts of second harmonic signals can be illustrated when dealing with atomic systems to examine the signal response of an electric field. The optical response of a medium excitation intensity is linearly behaved, while the low intensity of excitation also show linear relation between the electric field (\vec{E}) and induced dielectric polarization (\vec{P}) of incident radiations, as given in the following [9]:

$$\vec{P} = \varepsilon_0 X^{(1)} \vec{E} \qquad \dots (1)$$

where $X^{(1)}$ is commonly known as the linear susceptibility.

In nonlinear optics, the relationship between \vec{P} and \vec{E} could be generalised as the summation of a first order response $P^{(1)}$ and a series of nonlinear terms of increasing order to be given as:

$$\vec{P} = \vec{P}^{L} + \vec{P}^{NL} = \vec{P}^{(1)} + \vec{P}^{(2)} + \vec{P}^{(3)} + \cdots \qquad \dots (2)$$

where $\vec{P}^{(n)}$ is the n^{th} order nonlinear polarization [10].

By combining Equations (1) and (2), the nonlinear response of the material can be expressed as a Taylor expansion in terms of the applied electric field \vec{E} , as follows [11]:

$$P = \varepsilon_0 (X^{(1)} E_\omega + X^{(2)} E_\omega E_\omega + X^{(3)} E_\omega E_\omega E_\omega + \cdots) \qquad \dots (3)$$

where, the coefficients $X^{(n)}$ correspond to the tensor of the n^{th} -order nonlinear process. In order to obtain SHG, there must be three basic conditions: the existence of a high intensity light source such as laser, the principle of energy conservation, and the principle of momentum conservation. This depends on the process of phase matching. Since the photon energy is given as:

$$\begin{split} \mathbf{E} &= \mathbf{h}\omega = \mathbf{h}\mathbf{c}/\lambda & \dots (4) \\ \mathbf{c} &= \lambda\omega & \dots (5) \end{split}$$

where h is Blank constant, ω is the angular frequency, c is the speed of light, and λ is the wavelength. The physical origin to generate SHG is related to the nonlinear relationship [11] D

$$^{\rm NL} = 2 \varepsilon_0 \, \mathrm{X} \, \mathrm{d}_{\mathrm{eff}} \, \mathrm{E}^2 \qquad \dots (6)$$

where d_{eff} is a coefficient of a unit that is the inverse of the electric field unit (m/V). In the wave motion, the beats are generated by the movement of two waves of the same frequency and amplitude in the same region with opposite directions. The result of these two waves, by applying the principle of overlap, is twice the frequency and other is the frequency difference [12], where the electromagnetic wave with a fundamental frequency (ω) creates beats with itself to produce polarization of frequency (2ω) . To obtain this process, there is a condition where the speed of the phase of the polarized wave ($V_P = \frac{2\omega}{2K_{\omega}}$) is equal to the speed of the phase of the electromagnetic

wave $(V_E = \frac{2\omega}{K_{2\omega}})$, thus we can write this condition as [3]: K_2

$$\omega = 2K_{\omega} \qquad \dots (7)$$

and equation (7) can be written as:

$$n_{2\omega} = n_{\omega} \qquad \dots (8)$$

and the relation between the fundamental wave frequency (ω) and second harmonic wave frequency (ω_{SH}) is:

$$\omega_{SH} = 2 \omega \qquad \dots (9)$$

By multiplying the equations (7) and (9) with (\hbar) , one can obtain the following:

$$\hbar\omega_{\rm SH} = 2\hbar\omega \qquad \dots (10)$$

$$\hbar K_{2\omega} = 2\hbar K_{\omega} \qquad \dots (11)$$

where $\hbar\omega$ is photon momentum, and according to the law of energy conservation it must be [3]:

$$\frac{\mathrm{dI}_{2\omega}}{\mathrm{dz}} = -\frac{\mathrm{dI}_{\omega}}{\mathrm{dz}} \qquad \dots (12)$$

where I_{ω} and $I_{2\omega}$ represent the wave intensity at frequencies ω and 2ω , respectively. By applying equation (10), one can get:

$$\frac{\mathrm{dF}_{2\omega}}{\mathrm{dz}} = -\frac{2\mathrm{dF}_{\omega}}{\mathrm{dz}} \qquad \dots (13)$$

where F_{ω} and $F_{2\omega}$ are photon flux values of both waves. From equation (13), the SHG process produces a photon of frequency 2ω whenever two photons of frequency ω disappear. Equation (10) represents the principle of energy conservation, while equation (11) shows that the photon momentum is also conserved in this process.

1) Power Density and Conversion Efficiency

The processes of SHG by light waves falling at a frequency of ω_1 are described in the following two steps:

First Step: The resulting polarization wave from the SHG has a frequency of $2\omega_1$. The wavelength and phase velocity of this wave can be calculated in the medium by the refractive index of the fundamental wave n_1 , according to the relationship: [10]

$$\lambda_2 = \frac{c}{2\omega_1 n_1} \qquad \dots (14)$$

where *c* is speed of light, and λ_2 is wavelength of the fundamental wave.

Second Step: The energy moves from the polarization wave to the electromagnetic wave at a frequency of $2\omega_1$. The wavelength and phase velocity of the electromagnetic wave are generated by the refraction coefficient of the second harmonic wave (n₂), which can be calculated by the following relationship [11]:

$$\lambda_2 = \frac{c}{2\omega_1 n_2} \qquad \dots (15)$$

here λ_2 is the wavelength of the frequency doubling (SHG). This interaction is accompanied by a relatively high conversion efficiency.

2) Phase Mismatch

In order to obtain an efficient energy transmission, the two waves must be in the same phase. This leads to the concept $(n_1 = n_2)$. The phase mismatch between the polarized wave and the electromagnetic wave is usually expressed by the difference in the wave number (ΔK):

$$\Delta K = \frac{4\pi}{\lambda_1} (n_1 - n_2) \qquad \dots (16)$$

The conversion efficiency can be expressed as follows:

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$$\frac{P_{2\omega}}{P_{\omega}} = \tanh\left[IK^{1/2} \left(\frac{P_{\omega}}{A}\right)^{1/2} \frac{\sin\left(\frac{\Delta Kl}{2}\right)}{\left(\frac{\Delta Kl}{2}\right)}\right]^2 \qquad \dots (17)$$

The conversion efficiency is the ratio between the power generated from the SHG $(P_{2\omega})$ to the fundamental power (P_{ω}) , in which *l* is the coherent length. Since *K* is the coefficient of the material, it can be given as follows: [13]

$$\zeta = 2\eta^3 \omega_1^2 d_{\text{eff}}^2 \qquad \dots (18)$$

where η is the impedance of plane wave given in the following relation:

$$\eta = \left(\frac{\mu_0}{\varepsilon_0 \varepsilon_r}\right)^{\frac{1}{2}} \qquad \dots (19)$$

Equation (17) can be written as: [7]

$$\frac{P_{2\omega}}{P_{\omega}} = l^2 K \left(\frac{P_{\omega}}{A}\right) \frac{\sin^2(\frac{\Delta Kl}{2})}{(\frac{\Delta Kl}{2})^2} \qquad \dots (20)$$

where K is constant. Hence, the conversion efficiency depends on crystal length, power density, and phase mismatch.

2.3 Results and Discussion

In the practical phase, the setup of the proposed SHG experiment is established. Figure-1 pictures a captured photo for the practical setup of the proposed SHG experiment. It is obvious that there are six attenuators that are used to obtain different amounts of input laser power. This is useful to achieve different conversion efficiencies and make it possible to evaluate system performance of the proposed experiment. Whereas, a convex lens with 10cm focal length is placed in a distance (3.5-4 cm) from the crystal in order to achieve best focusing inside the crystal at the Riley region, i.e., least confusion area. This is very important to obtain SHG, while any focusing inside the crystal at a position that is not a Riley region will lead to a damage or self-scattering in the crystal, approaching 2-3 mm. Also, the numerical aperture of the lens and the incident spot are governed by ensuring that the laser ray do not touch the coating of the crystal, which may lead to fire the edges of the crystal damage. Therefore, aluminum foil is used at the front of the crystal to save it from the damage. The used nonlinear crystal is the BBO of $3 \times 3 \times 7$ mm size.



Figure 1-Schematic diagram of the proposed experiment setup.

The operation of the proposed experiment begins with switching-on the laser source and optimizing the alignment of both the pinhole and lens to make best focusing inside the crystal, which leads to both fundamental and second harmonic laser beams. These resulted beams will appear as much closer to each other. The double prism used after the crystal helps to diverge the second harmonic beam from the fundamental one. Then, each of them is detected using a specific powermeter. The fundamental beam is detected using a high powermeter, while the generated secondary harmonic beam, which is the output from the BBO crystal resulting from the SHG, is detected by a low powermeter. Figure-2 shows the spectrum of the fundamental beam converted from the SHG occurred in the used BBO nonlinear crystal. Figure-3 shows the spectrum of the second harmonic beam that is generated due to SHG occurrence, given by a spectrometer that is placed instead of the powermeter. The comparison between the two behaviors of the fundamental and the second harmonic beams can be deduced from Figures-(2 and 3). The intensity of the fundamental laser of 532nm wavelength is remarkably higher than that of the second harmonic generated laser beam of 266nm.



Figure 3-Spectrum of the generated secondary beam by SHG.

Conversion Efficiency Results

The conversion efficiency of the proposed SHG experiment was computed once for the simulated design using the theoretical concepts that are related to the optical parameters. Also, the conversion efficiency was measured another time when implementing the proposed SHG experiment practically, depending on the measurements of input and output powers. The following subsections explain more details about the results of efficiency for both theoretical and experimental methods.

1) Simulation Efficiency Results

MatLab program was implemented to compute the conversion efficiency for the proposed simulated SHG experiment design. The following strategy describes the main steps of the MatLab program.

1. Set θ =47.7°, φ =0°, d_{11} =2.32 *F/V*, d_{31} =0.116 *F/V*, d_{22} =0.1 *F/V*, (< d_{31}).

2. Set n_{01} =1.6734, n_{02} =1.7578, n_{e1} =1.5546, n_{e2} =1.6126.

3. Compute η (in Ω) using equation (19), where $\mu_0 = 1.256 \times 10^{-6} N/A$, $\varepsilon_0 = 8.854 \times 10^{-12} N/V$ and $\varepsilon_r = 6.7$.

4. Compute $\omega = c/\lambda$ (in H_z), where $c=3 \times 10^8 \text{m/s}$, and $\lambda = 0.532 \times 10^{-6} \text{m}$. Also, Compute $d_{eff} = d_{31} \times \sin(\theta) + (d_{11} \times \cos(3\varphi) - d_{22} \times \sin(3\varphi)) \times \cos(\varphi)$, compute *K* using equation (18), compute P_{out} using equation (20), where crystal length L=7mm, $A=1mm^2$, $P_{in}=$ (0-600) mW, and compute $Eff=(L^2 \times K \times (P_{in}/A)) \times 100\%$.

Figure-5 shows that the conversion efficiency of SHG is linearly proportional to the input power. The direct relation between them prove the slight effect of the input power (*Pin*) on the output efficiency. Whereas Figure-6 illustrates interesting logarithmic relations between the conversion efficiency and the output power, in which the efficiency is raising faster at low output values, then it reaches to the middle region where the progress of the efficiency slows down and few curved down, such that the increase of efficiency at lower *Pout* appears as being lower than that of the higher *Pin*.







Figure 6-Measured efficiency versus output power for 266 nm laser.

Figure-4 indicates that the output intensity power (*Pout*) is directly proportional to the input intensity (*Pin*), where the relation between them seems to be exponentially increasing along the allowed frequencies. The increase of *Pout* appeared as uniform, in which is the opposite to the increase in *Pin*. The slope of such raising behavior indicates reasonable efficiency, in which the input power leads to greater output power. **Practical Efficiency Results**

The use of the attenuators enables the decrease in the power of the input laser . The used attenuators are made of transparent glass of 3mm thickness and $2.5 \times 7.5 \text{mm}^2$ face area. Each attenuator can reduce 5% of the laser power beam. Therefore, six different input powers of laser beams are used to obtain six different output powers. The original input power of the 532nm laser source is specified to 600W, as written on the laser device, which is reduced by 5% each time the additional attenuator is added to the setup before the pinhole. The output power of both the fundamental and second harmonic beams/pulses is measured using powermeters. Table-1 lists different cases of input and output measurements that lead to different conversion efficiencies for the relations between the input, output, and efficiency for the considered laser source of the wavelength 532nm.

| No. of | Fundamental laser beam | | | Second Harmonic laser beam | | |
|-------------|------------------------|--------|--------|----------------------------|--------|--------|
| attenuators | Pin(mW) | Po(mW) | Eff. % | Pin(mW) | Po(mW) | Eff. % |
| 0 | 511 | 360 | 70.45 | 511 | 73.27 | 14.32 |
| 1 | 477 | 343 | 71.90 | 477 | 59.24 | 12.42 |
| 2 | 429 | 296 | 68.99 | 429 | 48.34 | 11.27 |
| 3 | 384 | 261.1 | 67.99 | 384 | 39.36 | 10.25 |
| 4 | 333 | 239.9 | 71.92 | 333 | 31.33 | 9.41 |
| 5 | 307 | 195.5 | 63.68 | 307 | 26.8 | 8.73 |
| 6 | 286 | 188.6 | 65.94 | 286 | 23.92 | 8.3 |

Table 1-Input-output powers and measured efficiency of 532nm laser.



Figure 7-Input power versus output power for 266nm laser and different number of attenuators.



Figure 8- Measured efficiency versus input power for 266nmlaser and different number of attenuators.



Figure 9- Measured efficiency versus output power for 266nm laser and different number of attenuators.

Figure-7 demonstrates that the output power of 266nm wavelength resulted from the SHG is increasing exponentially with the increase in the input power, where the output power is shown to be lower than the input power. Whereas, Figure (8) indicates a linear relationship between the conversion efficiency and the input power. The maximum obtained conversion efficiency was 14.32%, which is a well sufficient value according to the related literatures. In addition, Figure (9) indicates the relation between the conversion efficiency and the output power. The behavior of this relation appears as similar to that of the logarithmic curve which indicated that the maximum conversion efficiency was 12.97%.

Conclusions

A comparison was made between the simulation results as computed by MatLab program with the experimental results of the input laser of 532nm wavelength that was practically tested in the proposed SHG experiment. The results showed that the theoretical results are close to the experimental results. The input and output powers of the theoretical computations were 15.6% and 16%, respectively, which were close to the results obtained from the practical experiment that gave conversion efficiency values of about 14.32% and 12.97% for both input and output efficiencies, respectively. This refers to a low level of error percentage between the theoretical and experimental results. The application of Equation (2) on the achieved results showed that the estimated error rate is about 0.0452 for the input power, whereas the error rate between the theoretical and experimental readings is about 0.0991. In fact, these estimated errors are regarded as low values according to the related literatures. The results imply that the error in estimating the output power is greater than that in estimating the input power. This is due to a slight loss of laser beam throughout its interacting with the optical elements of the SHG experiment. Actually, the achieved results ensure the correct path of the research strategy and the acceptable results that can be safely applied to various fields.

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