Spontaneous Parametric Down-Conversion in a Beta Barium Borate Crystal

A Senior Project

By

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## Contents

1 Introduction ........................................ 4

2 Theory ........................................... 4
   2.1 Electromagnetic Waves ................................. 4
      2.1.1 Difference-Frequency Generation and Spontaneous Parametric Down-conversion 6
      2.1.2 Nonlinear Wave Equation ............................ 7
      2.1.3 Phase Matching .................................... 10
      2.1.4 DFG Wave Equations ................................. 10

3 Experiment ........................................ 12
   3.1 Equipment list ...................................... 12
      3.1.1 Laser ............................................. 12
      3.1.2 Crystal ............................................ 12
      3.1.3 Detectors .......................................... 13
   3.2 Electronics ......................................... 13
   3.3 Experiment Setup .................................... 13

4 Future Work ....................................... 14

### List of Tables

1 Type I vs Type II Phase Matching ........................ 10

### List of Figures

1 a) Illustration of DFG b) Energy Level Diagram .................. 7
2 a) Illustration Of Parametric DFG b) Parametric Energy Level Diagram ............. 11
3 Optical Path Block Diagram [5] ............................. 13
1 Introduction

This experiment was designed by Galvez et.al [1] in order to show undergraduate students the power of quantum optics. By building this experiment at California Polytechnic State University, we allow students to be exposed to graduate level concepts of nonlinear quantum optics in an undergraduate experiment. I was first exposed to these concepts in Glen D. Gillen’s Advanced Optics course and I wanted to continue working with these concepts and try to teach them to other students.

2 Theory

2.1 Electromagnetic Waves

Light is an electromagnetic wave which can be represented as a position and time-dependent complex wave with a time-dependent angular frequency of $\omega$. The position dependence is partially represented by a wave number $k$, or cycles per unit distance, which is defined by $k = n\omega/c$ where $n$ is the refractive index of the medium, defined as 1 in a vacuum, and $c$ is the speed of light in a vacuum. Being an electromagnetic wave, the light can be completely described by just its electric field given by [2]

$$\tilde{E}(t) = \tilde{E}_0 e^{-i\omega t} + c.c. ,$$  \hspace{1cm} (1)

where $\tilde{E}_0 = E_0 e^{i(k\cdot r + \delta)}$ is the complex amplitude of the wave, $E_0$ is the real amplitude of the wave, and $\delta$ is the phase constant.

Since light is a wave, if two waves interact in the same environment, the net wave will be the superposition of the two waves. So if the peaks of two waves both with amplitude $E_1$ add together, the observed wave will have amplitude $2E_1$, but if the peak of one wave with amplitude $E_1$ adds with the trough of the other wave with amplitude $E_1$, the observed wave will have an amplitude of 0. The Eq. 1 is given by [2]

$$\tilde{E}(t) = (\tilde{E}_1 + \tilde{E}_2) e^{-i\omega t} + c.c. .$$  \hspace{1cm} (2)

Equation 2 assumes that the two waves have the same frequency $\omega$ but if two waves have different frequencies, $\omega_1$ and $\omega_2$, Eq. 2 can be generalized to [2]

$$\tilde{E}(t) = \tilde{E}_1 e^{-i\omega_1 t} + \tilde{E}_2 e^{-i\omega_2 t} + c.c. .$$  \hspace{1cm} (3)
Light is a transverse wave, so the Electric field is perpendicular to the direction of motion which can be represented as $\mathbf{k}$, called the wavevector. This means that if the wavevector points in the $\hat{z}$ direction, the electric field can point in any direction of the $xy$ plane. This is called the polarization of the wave, as an $\tilde{\mathbf{E}}$ field oriented in the $\hat{z}$ direction would not interact with an $\tilde{\mathbf{E}}$ field oriented in the $\hat{y}$ direction. If an electric field is traveling through a medium, not a vacuum, the light may induce a polarization in the medium. The first order polarization of the medium can be described by the equation [2]

$$
\tilde{\mathbf{P}}(t) = \chi^{(1)} \tilde{\mathbf{E}}(t), \quad (4)
$$

where $\chi^{(1)}$ is the linear susceptibility, which is a scaler value. More generally, in nonlinear optics, Eq. 4 can be expanded using a Taylor series expansion, or [2]

$$
\tilde{\mathbf{P}}(t) = \chi^{(1)} \tilde{\mathbf{E}}(t) + \chi^{(2)} \tilde{\mathbf{E}}^2(t) + \chi^{(3)} \tilde{\mathbf{E}}^3(t) + \cdots \quad (5)
$$

$$
\equiv \tilde{\mathbf{P}}^{(1)}(t) + \tilde{\mathbf{P}}^{(2)}(t) + \tilde{\mathbf{P}}^{(3)}(t) + \cdots \quad (6)
$$

where $\chi^{(2)}$ and $\chi^{(3)}$ are the second- and third-order nonlinear optical susceptibilities, respectively. It is in these terms that all observable nonlinear effects are found. The second term of Eq. 6, the second-order component of the nonlinear polarization $\tilde{\mathbf{P}}^{(2)}(t)$, is given by the equation,

$$
\tilde{\mathbf{E}}^2(t) = E_1^2 e^{-2i\omega_1 t} + E_2^2 e^{-2i\omega_2 t} + 2E_1^* E_2 e^{-i(\omega_1 + \omega_2)t} + 2E_1 E_2^* e^{-i(\omega_1 - \omega_2)t} + 2E_1^* E_1 + 2E_2 E_2^* + \text{c.c.} \quad (7)
$$

This allows $\tilde{\mathbf{P}}^{(2)}(t)$ to written as [3]

$$
\tilde{\mathbf{P}}^{(2)}(t) = \chi^{(2)} \left[ E_1^2 e^{-2i\omega_1 t} + E_2^2 e^{-2i\omega_2 t} + 2E_1^* E_2 e^{-i(\omega_1 + \omega_2)t} + 2E_1 E_2^* e^{-i(\omega_1 - \omega_2)t} + \text{c.c.} \right] \quad (8)
$$

$$
+ 2\chi^{(2)} [E_1 E_1^* + E_2 E_2^*]
$$

The complex amplitude of $\tilde{\mathbf{P}}^{(2)}(t)$ can be written as a summation of the frequency components [3]

$$
\tilde{\mathbf{P}}^{(2)}(t) = \sum_n P(\omega_n) e^{-i\omega_n t} \quad (9)
$$

or in the specific case of two frequencies [3]
\[
\begin{align*}
P(2\omega_1) &= \chi^{(2)} E_1^2 & \text{(SHG)} \\
P(2\omega_2) &= \chi^{(2)} E_2^2 & \text{(SHG)} \\
P(\omega_1 + \omega_2) &= 2\chi^{(2)} E_1 E_2 & \text{(SFG)} \\
P(\omega_1 - \omega_2) &= 2\chi^{(2)} E_1^* E_2^* & \text{(DFG)} \\
P(0) &= 2\chi^{(2)} (E_1 E_1^* + E_2 E_2^*) & \text{(OR)}
\end{align*}
\]

Each expression has been labeled in the parentheses by the actual physical process it describes. Second harmonic generation, or SHG, is the process where two photons of the same frequency are absorbed and one photon is emitted with frequency \(\omega_3 = 2\omega_1\) or \(\omega_3 = 2\omega_2\). Sum frequency generation, or SFG, is a very similar process to SHG. Instead of two photons with the same frequency being absorbed, two photons with different frequencies are absorbed and a photon is emitted with frequency of \(\omega_3 = \omega_1 + \omega_2\). Difference frequency generation, or DFG, is analogous to SFG, but instead of a photon being emitted with a larger frequency, the photon emitted has a frequency of the difference of the photons, \(\omega_3 = \omega_1 - \omega_2\). DFG is covered in more detail in Secs. 2.1.1 and 2.1.4. Optical rectification, or OR, is a process which results in a static electric field within the medium.

It should also be noted that \(\chi\) is a tensor for a fixed geometry, so the polarization can also be written as

\[
P(\omega_3) = 4d_{\text{eff}} E(\omega_1) E(\omega_2)
\]

where \(d_{\text{eff}}\) is the effective value of a matrix. This allows you to express the polarization as a scalar value, which is quite valuable for calculations.

### 2.1.1 Difference-Frequency Generation and Spontaneous Parametric Down-conversion

In this paper we will be dealing with difference-frequency generation, which is given in Eq. 11 as \(P(\omega_1 - \omega_2) = 2\chi^{(2)} E_1 E_2^*\) which is also visualized in fig. 1. By conservation of energy, where the energy of each photon is \(h\omega\), we see that every photon generated at the difference frequency must have \(\omega_3 = \omega_1 - \omega_2\)

\[
\omega_3 = \omega_1 - \omega_2
\]
As we can see from fig. 1. and conservation of energy, whenever a photon is created at the difference-frequency, $\omega_3$, a photon at the higher frequency, $\omega_1$, must be destroyed and a photon at the lower frequency, $\omega_2$, must be created. Thus the $\omega_2$ field will be amplified by the down-conversion process. This process of using a second laser beam with frequency $\omega_2$ is called optical parametric down-conversion. If no pump beam is applied, two photon emission can still happen, but the efficiency of the down-conversion is much lower, and is called spontaneous parametric down-conversion and can only be achieved by properly tuning your crystal.

Optical parametric down-conversion can be used to create an amplifier by placing highly reflective mirrors on either side of the nonlinear medium, the pump beam will continuously stimulate the down-conversion and amplify the intensity of both $\omega_2$ and $\omega_3$.

### 2.1.2 Nonlinear Wave Equation

To derive the nonlinear wave equation we start with Maxwell’s equations in matter: [3]

\[
\nabla \cdot \tilde{D} = \tilde{\rho} \tag{12}
\]

\[
\nabla \cdot \tilde{B} = 0 \tag{13}
\]

\[
\nabla \times \tilde{E} = \frac{\partial \tilde{B}}{\partial t} \tag{14}
\]

\[
\nabla \times \tilde{H} = \frac{\partial \tilde{D}}{\partial t} + \tilde{J} \tag{15}
\]

Figure 1: a) Illustration of DFG b) Energy Level Diagram
Since we are using waves in nonconducting material, we may assume that there are no free charges ($\dot{\rho} = 0$) or free currents ($\dot{\mathbf{J}} = 0$). We can also assume that the material is non-magnetic ($\mathbf{B} = \mu_0 \mathbf{H}$) and that $\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P}$. By taking the curl of Eq. 14 we find that the nonlinear wave equation takes the form of [3]

$$\nabla^2 \mathbf{E} - \frac{\epsilon^{(1)}(\omega_n)}{c^2} \frac{\partial^2 \mathbf{E}_n}{\partial t^2} = \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 \mathbf{P}^{NL}_n}{\partial t^2}. \quad (16)$$

By solving Eq. 16 for a plane wave with angular frequency $\omega_3$ that propagates in the $+z$ direction in the absence of a nonlinear sourcing term we find [3]

$$\mathbf{E}_3(z, t) = A_3 e^{i(k_3 z - \omega_3 t)} + \text{c.c.}, \quad (17)$$

where

$$k_3 = \frac{n_3 \omega_3}{c}, \quad n_3 = \left[\epsilon^{(1)}(\omega_3)\right]^{1/2}, \quad (18)$$

and $A_3$ is a constant.

By applying the assumption of Eq. 9 the first order approximation polarization given in Eq. 6 can be written as [3]

$$\mathbf{P}_3(z, t) = P_3 e^{-i\omega_3 t} + \text{c.c.}. \quad (19)$$

Equation 17 can be generalized to [3]

$$
E_i = A_i e^{i(k_i z - \omega_i t)}.
$$

Equation 19 can be written by substituting Eq. 20 into Eq. 11 to yield [3]

$$\mathbf{P}_3 = 4d_{eff} A_1 A_2 e^{i[(k_1 + k_2)z - (\omega_1 + \omega_2)t]}.$$  

(21)

Substitution of Eqs. 17 and 21 into Eq. 16 and changing $\nabla^2$ into $\frac{\partial^2}{\partial z^2}$ we get [3]

$$\frac{d^2 A_3}{dz^2} + 2i k_3 \frac{dA_3}{dz} = \frac{-16\pi d_{eff} \omega_3^2}{c^2} A_1 A_2 e^{i\Delta k z}, \quad (22)$$

where $\Delta k$ is called the wavevector mismatch, or [3]

$$\Delta k = k_3 - k_1 - k_2. \quad (23)$$

Since the amplitude $A_3$ is slowly varying, it can be assumed that [3]

$$\left| \frac{d^2 A_3}{dz^2} \right| \ll \left| \frac{dA_3}{dz} \right|. \quad (24)$$
Now Eq. 22 can be written as [3]

\[
\frac{dA_3}{dz} = \frac{8\pi i d_{\text{eff}} \omega_3^2}{k_3 c^2} A_1 A_2 e^{i\Delta k z}.
\] (25)

By using a very similar process we find that [3]

\[
\frac{dA_1}{dz} = \frac{8\pi i d_{\text{eff}} \omega_1^2}{k_1 c^2} A_3 A_2^* e^{i\Delta k z},
\] (26)

and

\[
\frac{dA_2}{dz} = \frac{8\pi i d_{\text{eff}} \omega_2^2}{k_2 c^2} A_3 A_1^* e^{i\Delta k z}.
\] (27)

The condition for perfect phase matching is a wavevector mismatch of zero, or [3]

\[\Delta k = 0.\] (28)

Phase matching is explained in more detail Sec 2.1.3. By satisfying the condition of Eq. 28, we find that [3]

\[A_3(L) = \frac{8\pi i d_{\text{eff}} \omega_3^2}{k_3 c^2} A_1 A_2 \left(\frac{e^{i\Delta k L} - 1}{i \Delta k}\right).\] (29)

This leads to the an intensity of [3]

\[I_3 = \frac{32\pi d_{\text{eff}}^2 \omega_3^4 |A_1|^2 |A_2|^2 n_3^2}{k_3^2 c^3} \left| e^{i\Delta k L} - 1 \right|^2,\] (30)

and, by definition of the square modulus, the last part of \(I_3\) becomes [3]

\[\left| e^{i\Delta k L} - 1 \right|^2 = L^2 \text{sinc}^2 (\Delta k L/2).\] (31)

By solving for \(I_1\) and \(I_2\) and plugging them, along with Eq. 31 into Eq. 29 we find [3]

\[I_3 = \frac{512\pi^5 d_{\text{eff}}^2 I_1 I_2}{n_1 n_2 n_3^3 \lambda_3^2 c^6} L^2 \text{sinc}^2 (\Delta k L/2).\] (32)

Eq. 32 clearly shows the intensity follows a \(\text{sinc}^2\) that is dependent upon \(L\) which is the length of the nonlinear medium. This sinc relationship shows a strong dependence upon the wavevector mismatch, and highlights the importance of perfect phase matching (Eq. 28).
2.1.3 Phase Matching

When looking at the intensity equation Eq. 32, we can clearly see that the intensity reaches a maximum as $\Delta k = 0$, where $\Delta k$ satisfies Eq. 23.

For a birefringent crystal, there are two axes, the fast and slow axis. In nonlinear optics these are called the extraordinary index $n_e$ and the ordinary index $n_o$. Where the extraordinary index is along the polarization axis of the crystal and the ordinary axis is perpendicular to the polarization axis.

There are two possible ways for the light to enter and potentially be phase matched. When the two lower-frequency waves have the same polarization, we define that as type I phase matching. Type II phase matching is when the two polarizations are orthogonal. Typically type I phase matching is easier to achieve. This leads to classifications for crystals shown in table 1.

<table>
<thead>
<tr>
<th>Type</th>
<th>Positive uniaxial ($n_e &gt; n_o$)</th>
<th>Negative uniaxial ($n_e &gt; n_o$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Type I</td>
<td>$n^2_3\omega_3 = n^2_1\omega_1 + n^2_2\omega_2$</td>
<td>$n^2_3\omega_3 = n^2_1\omega_1 + n^2_2\omega_2$</td>
</tr>
<tr>
<td>Type II</td>
<td>$n^2_3\omega_3 = n^2_1\omega_1 + n^2_o\omega_2$</td>
<td>$n^2_3\omega_3 = n^2_1\omega_1 + n^2_o\omega_2$</td>
</tr>
</tbody>
</table>

Table 1: Type I vs Type II Phase Matching

There are two ways to tune a birefringent crystal, angle tuning and temperature tuning. Temperature tuning is useful when the crystal you are using has an index of refraction that is highly temperature dependent. For most crystals, angle tuning is more practical.

It can be shown that the effective refractive index $n_e(\theta)^2$ is given by [3]

$$\frac{1}{n_e(\theta)^2} = \frac{\sin^2(\theta)}{n^2_e} + \frac{\cos^2(\theta)}{n^2_o},$$

where $n^2_e$ is the principal value of the extraordinary refractive index. It is clear that $n_e(\theta)^2$ is equal to the principal value at $\theta = 90$ degrees. Phase matching occurs where $n_e(\theta)^2$ satisfies the condition $\Delta k = 0$.

2.1.4 DFG Wave Equations

By solving Eqs. 26 and 27 for perfect phase matching ($\Delta k = 0$), and differentiating with respect to $z$ we find that [3]

$$\frac{d^2 A_2}{dz^2} = \frac{64\pi^2 \omega_1^2 \omega_2^2 d^2_{eff}}{k_1 k_2 e^4} A_3 A_3^* A_2 \equiv \kappa^2 A_2,$$

where $d^2_{eff}$ is the effective nonlinear coefficient.
where $\kappa$ is defined as the coupling constant [3]

$$\kappa^2 = \frac{64\pi^2 \omega_1^2 \omega_2^2 d_{eff}^2}{k_1 k_2 c^4} |A_3|^2 . \quad (35)$$

The general solution to this differential equation is of the form [3]

$$A_3 (z) = C \sinh (\kappa z) + D \cosh (\kappa z) . \quad (36)$$

By applying boundary conditions of [3]

$$A_2(0) = 0, A_1(0) = \text{const.}$$

the solution to Eqs. 26 and 27 becomes [3]

$$A_1(z) = A_1(0) \cosh (\kappa z) \quad (37)$$
$$A_2(z) = i \left( \frac{n_1 \omega_2}{n_2 \omega_1} \right)^{1/2} \frac{A_3}{|A_3|} A_1^*(0) \sinh (\kappa z) . \quad (38)$$

When applying the condition of equal frequencies for $\omega_1$ and $\omega_2$ and conservation of energy we get

$$\omega_1 = \omega_2 = \frac{\omega_3}{2} , \quad (39)$$

as illustrated in fig. 2. When phase matching for $\omega_1 = \omega_2$ and $\omega_3$ is achieved spontaneous parametric down-conversion becomes possible, and we find that Eqs. 34 becomes

$$\frac{d^2 A_1}{dz^2} = \frac{64\pi^2 \omega_1^4 d_{eff}^2}{k_1 k_2 c^4} A_3^* A_3 A_1 \equiv \kappa^2 A_1 , \quad (40)$$
where the coupling constant is now described by

\[ \kappa^2 = \frac{64\pi^2}{k_1^2 c^4} \omega_1^4 |A_3|^2. \]  

(41)

3 Experiment

3.1 Equipment list

3.1.1 Laser

There were two lasers in our set up as illustrated in fig. 3, the 405 nm diode laser, which is used as the beam that interacts with the crystal and acts as \( \omega_3 \) and the He-Ne laser which acts as an alignment beam as the He-Ne is much easier to see than the difficult to see diode laser. To ensure that the lasers are traveling the same path we placed two irises in the beam path. In order to control the polarization and intensity we cross two polarizers across the crystal, so only the down-converted photons that change polarization will be let into the interferometer.

3.1.2 Crystal

For this experiment we used a Beta-Barium Borate crystal (BBO crystal), which is a negative uniaxial crystal, that was cut for type I phase matching. By using values given by \[4\]

\[ d_{eff} = d_{31} \sin(\theta) + (d_{11} \cos(3\phi) - d_{22} \sin(3\phi)) \cos(\theta) \]  

(42)

and \( n_o \) and \( n_e \) given by Galvez et.al \[1\]

\[ n_e^2 = 2.3753 + \frac{0.01224}{\lambda^2 - 0.01667} - 0.01516\lambda^2 \]  

(43)

\[ n_o^2 = 2.7359 + \frac{0.01878}{\lambda^2 - 0.01822} - 0.01354\lambda^2 \]  

(44)

which leads to values of

\[ n_e (\lambda = 810\text{nm}) = 1.5442 \]  

(45)

\[ n_o (\lambda = 810\text{nm}) = 1.6603 \]  

(46)

for our BBO crystal at 810-nm wavelength.
3.1.3 Detectors

In order to detect the individual photons we used two Perkin-Elmer Avalanche Photodiodes (SPCM-AQRH-13) (APD). APD’s were chosen due to their high quantum efficiency at the desired 810-nm wavelength. The APD’s are incredibly sensitive to photons and if the concentration of photons is too large the APD can be permanently damaged. In order to prevent this we performed the experiment in complete darkness and constructed a black box to prevent excess photons from entering the detectors.

3.2 Electronics

To ensure that the detected pulses were recorded, we set up a nuclear box with two Single Channel Analyzers (Ortec 406A) to convert the analog pulses to digital pulses. We then passed them through a Fast Coincidence Module (Ortec 414A) to ensure that any pulses are detected only if each detector is struck with a photon simultaneously. We then sent the signal into a Counter (Ortec 994) to display the number of counts.

3.3 Experiment Setup

![Optical Path Block Diagram](image)

Figure 3: Optical Path Block Diagram [5]
Figure 3 shows how we arranged everything in order to run the experiment. We set everything onto a \(2' \times 3'\) optical breadboard, so that the experiment would be easily transportable.

4 Future Work

This project still has much work to do. Due to time constraints, phase matching the BBO crystal was not achieved, a necessary set to record meaningful data. Once the apparatus is perfectly aligned, the next set will be to automate the data taking process, which would involve using a LabVIEW enabled computer to control both the arm length of the interferometer and to record the data from the detectors.

Once this is completed, undergraduate students will be able to preform experiments that will be able to prove quantum entanglement in photons and allow students to prove that a photon exists.

References


