Second Harmonic Generation
in Nonlinear Optical Crystal

Diana Jeong

1. Introduction

In traditional electromagnetism textbooks, polarization in the dielectric material is linearly proportional to the applied electric field. However since in 1960, when the coherent high intensity light source became available, people realized that the linearity is only an approximation. Instead, the polarization can be expanded in terms of applied electric field. (Component - wise expansion)

$$(1) \qquad P_k = \varepsilon_0 (\chi_{ik}^{(1)} E_i + \chi_{ijk}^{(2)} E_i E_j + \chi_{ijkl}^{(3)} E_i E_j E_k + \cdots)$$

Other quantities like refractive index (n) can be expanded in terms of electric field as well. And the non linear terms like second (E^2) or third (E^3) order terms become important. In this project, the optical nonlinearity is present in both the source of the laser-mode-locked laser- and the sample.

Second Harmonic Generation (SHG) is a coherent optical process of radiation of dipoles in the material, dependent on the second term of the expansion of polarization. The dipoles are oscillated with the applied electric field of frequency w, and it radiates electric field of 2w as well as 1w. So the near infrared input light comes out as near UV light.

In centrosymmetric materials, SHG cannot be demonstrated, because of the inversion symmetries in polarization and electric field. The only odd terms survive, thus the second order harmonics are not present.

SHG can be useful in imaging biological materials. For example, the collagen fibers and peripheral nerves are good SHG generating materials. Since the SHG is a coherent process it, the molecules, or the dipoles are not excited in terms of the energy levels. Thus SHG doesn't, in principle, photodamage the cells because there are no photo excitations. Also SHG is from the intrinsic property of the molecules and the input light, so external dyes are not necessary.

The goal of this project, for now, is to investigate the SHG signal quantitatively. Particularly, to explore angular, spatial, intensity dependence of SHG signals from βBaBO4 (BBO) crystal. This project is a little stepping stone for SHG of the biological imaging in the near future.

2. Theory

1) Radiation of the Dipoles

The single BBO molecule can be regarded as a dipole moment.

(2)
$$\vec{\mu}_{2\omega} = \frac{1}{2}\beta E_{\omega}^2 \hat{z}$$

The electric field and radiated power from the dipoles are following

(3)
$$\bar{E}_{2\omega}(\psi) = -\frac{\mu_{2\omega}\omega^2}{\pi\varepsilon_0 c^2 r}Sin(\psi)\exp(-2i\omega[t])\hat{\psi}$$

(4)
$$P_{2\omega}(\psi) = \frac{3}{16\pi}\sigma_{SHG}Sin^2(\psi)I_{\omega}^2, \sigma_{SHG} = \frac{4n_{2\omega}\hbar\omega^5}{3\pi n_{\omega}^2 \varepsilon_0^3 c^5}|\beta|^2$$

with the cross-section for the SHG.

Integration over whole solid angle gives

.

(5)
$$P_{SHG} = \frac{1}{2}\sigma_{SHG}I_{\omega}^2$$

Like Two photon fluorescence, the intensity depends on the square of the applied field but the hyperpolarizability plays a role here. The SHG signal depends on the square of the magnitude of the hyperpolarizability, but the Two- photon signal depend on the imaginary part of hyperpolarizability. So the signal of SHG tends to be much smaller than Two-photon signal.

2) Phase Matching

The phase matching of the radiated electric field of w and 2w is critical in SHG. They have to be in phase, so as not to interfere destructively. The applied electric field is strong enough to generate the second order radiation, but not so strong so that the dominant term is the first order term. So we could think of SHG signal as a perturbation and thus those lights of two frequencies should be added, and should not cancel each other out.
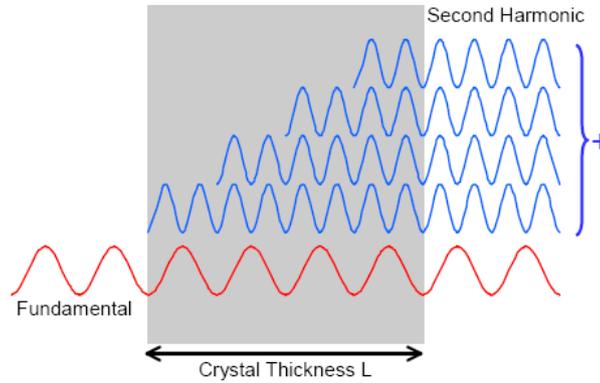
Fig1. Concept of phase matching [1]

The total output is sum, or the integration of the all dipoles at every position in the material.

(6)
$$\eta = \frac{1}{L^2}\left|\int_0^L e^{(i2k_\omega z)}e^{(-ik_{2\omega}z)}dz\right|^2 = \frac{1}{L^2}\left|\int_0^L e^{(i\Delta kz)}dz\right|^2 = \left(\frac{Sin(\Delta kL/2)}{\Delta kL/2}\right)^2$$

So the maximum is achieved when the phases are the same. This means the lights should travel the medium at same velocities. The refractive index for the two different frequencies should be same. It brings up a tricky issue. The index of refraction is

(7)
$$n \propto \sqrt{\varepsilon}$$

And the permittivity in simple electron model is

(8)
$$\varepsilon(\omega) = 1 + \frac{4\pi n e^2}{m}\sum_j \frac{f_j}{-\omega^2 + \omega_j^2 - i\omega\gamma_j}$$

Thus, as wavelength increases the index of refraction decreases. To create the SHG signal, we really need to figure out ways to compensation in refractive index to match phases.

3) SHG Samples- Birefringence in BBO

There are few ways to get away with this dispersion in refractive index.

a. The lipid bilayer with one leaflet dyed is a good example. Only the layer of dyes will radiate while the SHG from the lipid layers will cancel each other out. And since the dye molecules and lipid molecules will bind in certain orientation only, every molecule is aligned. But the biggest problem is making continuous, homogenous lipid layer is very difficult and tedious.

b. The collagen fiber is also a good sample. The collagen protein is abundant in animal body. The collagen should be in form of fiber, because of the inversion symmetry and thickness. Collagen fibers are usually few microns and within few microns range, the coherence in

the phase is kept. So collagen fiber is ideal for SHG signal, and to obtain image from it. Collagen fibers can be easily obtained from rat tail tendon or peripheral nerves. But collagen fibers are sensitive to the moisture, and in this optical setup it was hard to maintain suitable moisture.

c. The most accessible sample was birefringent crystals. Birefringence is a phenomenon arises from the anisotropy of the crystal structure. In microscopic sense, different molecules radiate in different ways and this leads to difference in refractive index.

Fig 2. Concept of Birefringence [5]

BBO is a birefringent material and the refractive index of extraordinary axis is smaller than that of ordinary axis. The indices are dependent on both the frequency of light and the angle between propagation and optical axis.

(9)
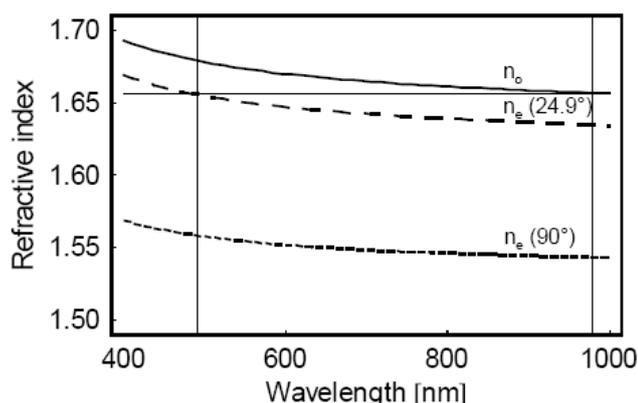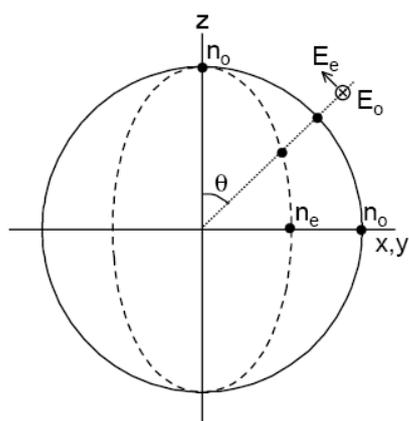$$\frac{1}{n_e(\Theta, \lambda)} = \frac{Sin\Theta^2}{n_e(\lambda)^2} + \frac{Cos\Theta^2}{n_0(\lambda)^2}$$

Fig 3.and Fig 4. Angle and Wavelength Dependence of Refractive index of BBO [1]

The goal is to find angle that n_2w and n_w are the same. At angle 65.1, both of them are 1.657.

In professor Kleinfeld's lab there was BBO crystal available so in this project I used BBO crystal.

4) SHG of focused beams-Gaussian Beam

In this project, the plane wave laser light is expanded and focused by objective. Thus it is necessary to take Gaussian Beam into consideration rather than plane wave.
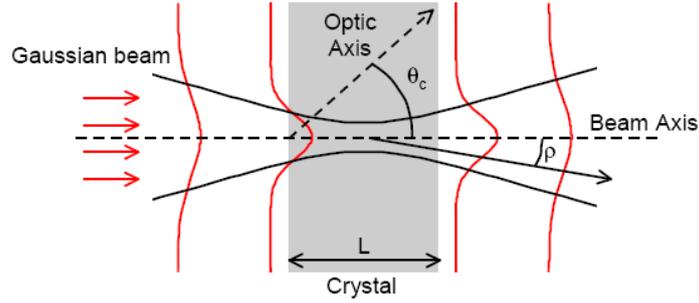

Fig 5. Profile of Gaussian Beam [1]

First, we should examine the propagation of nonlinear polarization. Everything starts from Maxwell's equation

(10)
$$\nabla^2 \vec{E}(\vec{r},t) + \frac{n^2}{c^2}\frac{\partial^2}{\partial^2 t}\vec{E}(\vec{r},t) = -\mu_0 \frac{\partial^2}{\partial^2 t}\vec{P}(\vec{r},t)$$

Using the expansion given in the beginning, second harmonic component is given

(11)
$$\nabla^2 \vec{E}_2(\vec{r}) + \frac{(2\omega)^2}{c^2}\vec{E}_2(\vec{r}) = \frac{(2\omega)^2}{c^2}\vec{P}(\vec{r})$$

With assumption of $\left|\nabla A\right| << k_z \left|A\right|$

(12)
$$E(\vec{r}) \equiv \vec{A}(\vec{r})e^{ik_2 z}, \frac{\partial}{\partial z}A(\vec{r}) = \frac{i\omega}{2nc}P_2(\vec{r})e^{-ik_2 z}$$

The last equation can be examined in two ways. First, in the limit of the plane wave approximation,

(13)
$$I_{2\omega} = C^2 I_\omega^2 \frac{L^2}{\pi \omega_0^2}$$

Now, the Gaussian beam is expressed following.

(14)
$$E_1(\vec{r}) = \frac{E}{1+i\tau} \exp(-\frac{x'^2+y'^2}{\omega_0^2(1+i\tau)}) \exp(ik_1 z)$$

There is no simple analytic solution for real Gaussian beam except numerically plotted graph.

(15)

$$I_{2\omega} = C^2 I_\omega^2 \frac{Lb}{\pi\omega_0^2} h(B,\xi)$$

$$b \equiv \omega_0^2 k_1 \equiv \omega_0^2 n_1 \frac{2\pi}{\lambda_1}$$

$$\xi \equiv \frac{L}{b}, B \equiv \frac{1}{2}\rho\sqrt{k_1 L}$$
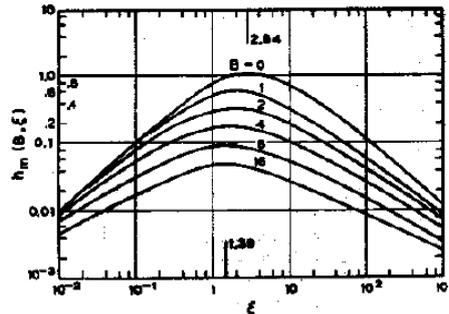
the ρ is walk-off angle of BBO, 60mrad.

Fig 6. Numerical evaluation of function h [2]

3. Experimental Setup

1) Mode-locked laser

As we have seen above, to achieve the SHG we need a light source of very high power. The power that nonlinearity of optical property of the sample can be manifested is few miliwatts laser. However, to achieve that high intensity laser output in continuous wave (cw) mode, we need exceedingly high energy. Instead, we seek for pulse laser.

To make a pulse we first need resonant cavity. In resonant cavity only certain modes are allowed due to the boundary conditions.

(16)
$$\lambda m = \frac{L}{2}, f_m = \frac{mc}{2L}$$

And if the phases of the each modes are locked, i.e. mode-locked, the modes can be added up. If not there will be destructive interferences and mode-locking will not be achieved.

(17)
$$U(x,t) = A_0 e^{i2\pi f_0 (t-\frac{x}{c})} + A_1 e^{i2\pi f_1 (t-\frac{x}{c})} + \cdots = \sum_q A_q e^{i2\pi f_q (t-\frac{x}{c})}$$

(18)
$$I(t,x) = |A|^2 \frac{Sin^2(m\frac{\pi c}{2L}(t-\frac{x}{c}))}{Sin^2(\frac{\pi c}{2L}(t-\frac{x}{c}))}$$

Due to the uncertainty principle, the shorter the delta t gets, the bigger the delta w will get. So if we succeed in adding up all the modes together, we get the train of pulses.
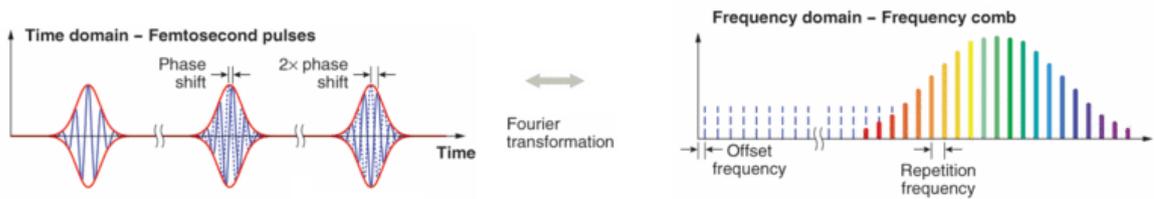

Fig7. Concept of Modelocking in Fourier Transform [6]

The technical difficulty arises in mode-locking, since the phases of all the modes should be the same. This can be done by utilizing nonlinearity in refractive index, the Kerr effect. In high intensity electric field, the refractive index varies as an expansion of the electric field,

$\Delta n = \lambda K E^2$, K Kerr constant, λ wavelength of light, E magnitude of electric field.

Since the spectrum of the laser will be Lorenztian distribution, there will be different optical paths for different intensity, thus the paths will vary due to different frequencies. So the phases of the various modes can be adjusted for the modelocking to be achieved.

Detailed calculation for the condition of the modelocking can be done by Jones matrices.

In this setup I have used Clark-MXR MAGELLAN femto-second mode-locking laser. The specification of the laser is following.

Fig 8. Picture of uncovered Clark MXR Modelocked laser [7]

By adjusting the waveplates and fine adjustment, the modelocking and maximum intensity can be achieved.

To verify if the laser is mode locked, I have checked intensity with powermeter, spectrum with spectronometer and signal from photodiode. Once the laser is mode locked I was able to see the broad spectrum around 1030nm. However the maximum power of modelocked laser was only 18mw, only about 1/3 of the expected value.
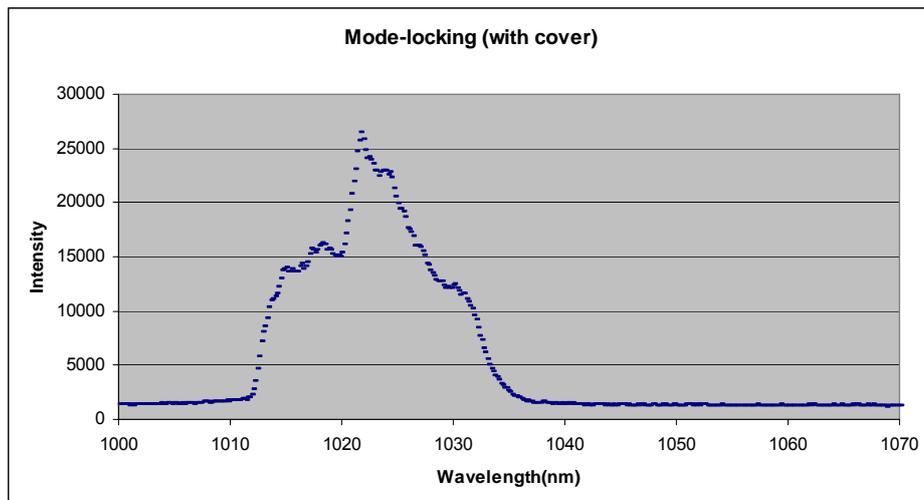

Fig 9. Output spectrum of Modelocked laser

And unfortunately, in the 9th week, the laser completely went out of alignment. So I had to move everything to Professor Kleinfeld's lab and set things up again. In real measurements, I did not use Clark laser.

2) Optical Setup

The optical setup consists of Neutral Density filter wheel, beam expander, objective, (collecting lens), filters and PMT. The Neutral Density filter wheel (ND filter) was used for varying intensity and prevention of overload of the PMT.
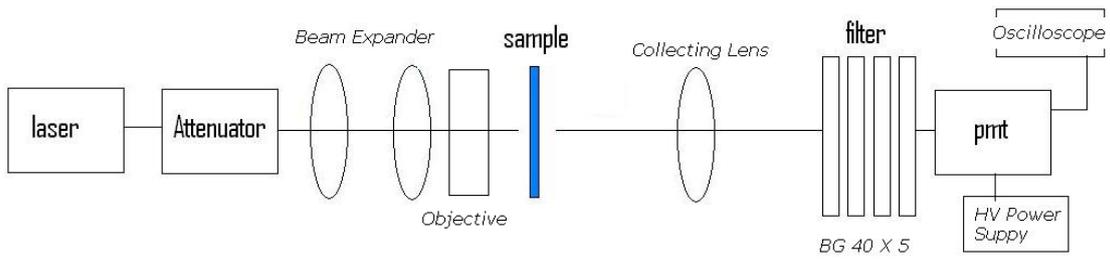
Fig10. Schematic of Experimental Setup

The purpose of the beam expander is to enlarge the size of the beam to fully use the high NA (numerical aperture) lens for focusing. Focusing is critical step to enhance the intensity further on. Here I have used Beam expander composed of one diverging lens and one converging lens after. The magnification is $M_0 = \dfrac{f_d}{f_c}$. The distance between the lenses should be $f_c - f_d$.
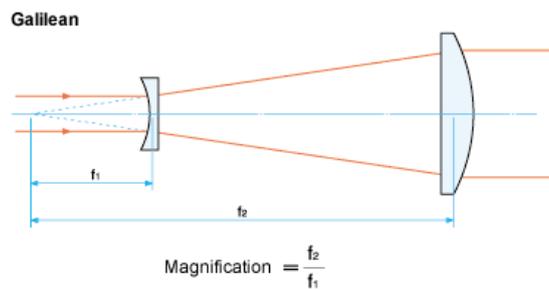

Fig 11. Concept of Galilean Beam expander [8]

The PMT was used to amplify the intensity of the output light. Even the small amount of photons will cause photoelectric effect and overload the PMT. So I have used five Schott BG40 filters to prevent the input laser light getting into PMT.
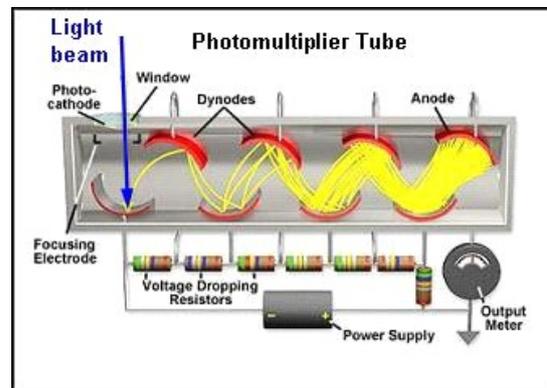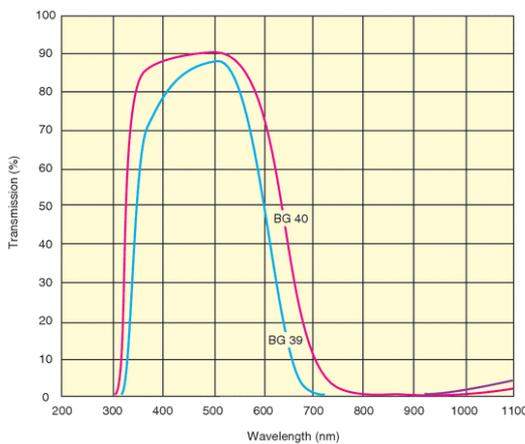

Fig 12. Specification of BG39 and BG40 filters [9] and Fig 13.Concept of PMT [10]
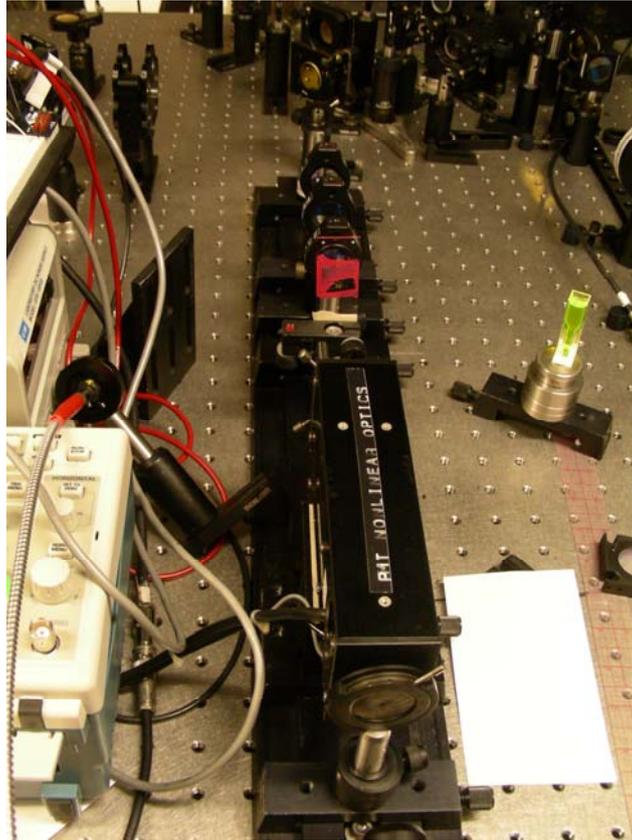
The Actual setup looked like this.

Fig. 14. Optical Setup in Kleinfeld's lab. The Green fluorescence cuvet was used for two photon signal to verify if we had enough laser power.

4. Data

There is major problem in measuring spectrum of the SHG signal due to the change in the light source. With the Clark MXR mode-locked laser of wavelength 1030 nm, I would have gotten 515nm signal. But the new light source has wavelength of 800nm, which leads to the 400nm signal from BBO. The spectronometer that I used had wavelength range from 470nm to 1150nm. So I did see a violet colored light from the crystal, I had no way of verifying if it is real SHG or not except the filter. Since the minimum wavelength of spectronometer is 470nm and the filter only allows light from 300nm to 600nm, the estimated range of SHG wavelength is 300-470nm.

I changed ND values of the attenuator to measure the input intensity dependence of SHG signal.
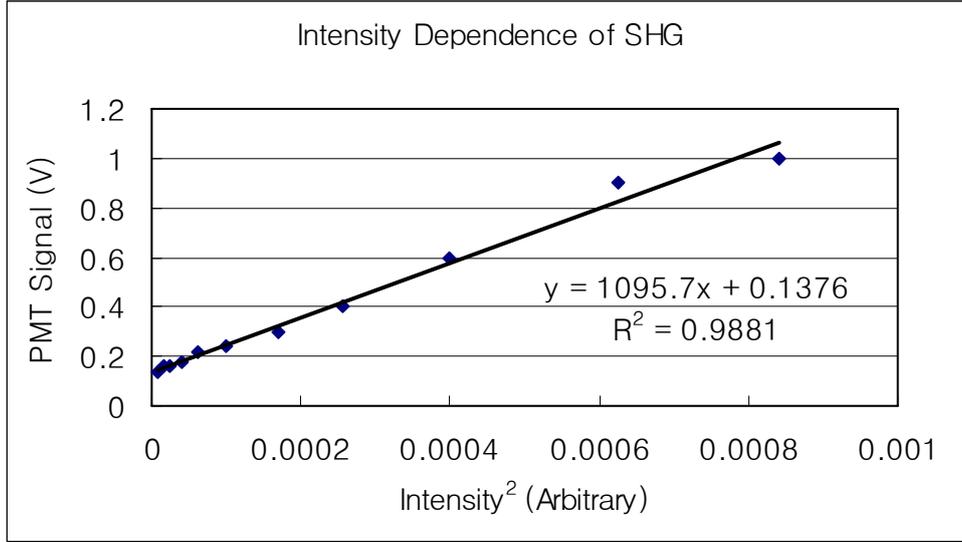
(19)
$$P_{in}10^{-ND} = P_{out}$$

Fig15. Intensity Dependence of SHG

In the first part of the theory there is a formula (4) for intensity versus power radiated.

$$(20) \qquad P_{2\omega}(\psi) = \frac{3}{16\pi}\sigma_{SHG}Sin^2(\psi)I_\omega^2 , \sigma_{SHG} = \frac{4n_{2\omega}\hbar\omega^5}{3\pi n_\omega^2 \varepsilon^3{}_0 c^5}|\beta|^2$$

With this formula and n_2w~n_w, we can figure out the hyperpolarizability of the BBO crystal.

I managed to get quadratic dependence of the SHG signal on intensity, but I need two more information to fully analyze data. I need to figure out input intensity of the laser and conversion rate of the PMT.

I also observed there is a sharp sensitivity in incident angle. But I didn't have a setup to sensitively change the angle of the crystal to quantitize the sensitivity and compare it with the theoretical value. I would need automated rotating stage. Another observation made was sensitivity of the position of the BBO crystal. I would also need to move the crystal and scan through the optical axis.

5. Further experiment

As I mentioned above, due to the short of time, I only managed to observe the angular and position dependence of the SHG signal qualitatively. Along with figuring out the hyperpolarizability, quantitative measurement of the dependencies should be made. These will be the basic steps. In order to do so, I would need automated rotating stage and automated position scanning system.

Another direction of the further experiment should focus on various samples of the SHG. To image biological systems, understanding angle dependence and focus dependence is

important. We need to first figure out optimized condition for imaging and then try to image the biological samples. And imaging peripheral nerve with SHG will be exciting challenge.

\<Reference\>

(1) Loaiza and Dood, *Second-Harmonic Generation*, Leiden University

(2) G.D Boyd, A. Ashkin, J.M. Dziedzic and D.A. Kleinman, Phys. Rev. **137**,A1305(1965)

(3) L. Moreaux, O. Sandre and J. Mertz, J.Opt.Soc.Am.B, **17**, 1685(2000)

(4) S.E. Skipetrov, Disorder is New Order, Nature **432**, 285, (2004)

(5) Optical Birefringence, micro.magnet.fsu.edu

(6) FFT of Modelocked laser,www.laserfocusworld.com

(7) Clark MXR, correspondence

(8) Beam expanders, www.sigma-koki.com

(9) BG 39,40 product specification,   http://www.thorlabs.com

(10) PMT, www.nt.ntnu.no