Lab Report Nonlinear Optics

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I. INTRODUCTION

Nonlinear optics has become a important field in photonics since the discovery of the Laser in 1960 allowed us to study this effect. This new light source allowed us to produce intensities which had not been possible before, nonlinear effects require high intensity light to interact with the medium. With this we make a system that generates photons frequency doubled or tripled the incoming light. In this experiment we're interested in investigate the frequency doubling and tripling phenomenon. This is a very interesting phenomenon since it allows us to tailor a light source as we wish with out all the constraints that come with making a laser system of some hard to achieve frequency.Thus, second and third harmonic generation is a technique that gives us access producing intense light sources in spectral regions previously unavailable by classic linear optics.

II. THEORY

To produce this interaction between light and the electric dipole moment of a material requires the conservation off energy and momentum which are known as the phase matching condition, this can be set for second harmonic generation (SHG). The phase mismatch :

$$\Delta k = k_{\mathrm{f}} - 2k_{\mathrm{sh}} \tag{1}$$

where k_{f}, $2k_{\mathrm{sh}}$ are the wavenumbers of the fundamental and second-harmonic beam, respectively. Lets consider the case with zero mismatch $k_{\mathrm{f}} = 2k_{\mathrm{sh}}$ this can only be achieved if the refractive index fulfill:

$$n_\omega = n_{2\omega} \tag{2}$$

One can consider non-critical phase matching which means that the temperature of the non-linear medium is varied to fulfill the phase-matching condition. Or critical phase-matching where the angle dependence of the refractive index of uni-axial crystals, is used to achieve phase matching. The crystal is cut such that the transmission is maximised.
The critical phase matching angle can be computed the following formula [1]:

$$\sin^2\theta_{\mathrm{m}} = \frac{n_{\mathrm{o}}^{-2}(\omega) - n_{\mathrm{o}}^{-2}(2\omega)}{n_{\mathrm{e}}^{-2}(2\omega) - n_{\mathrm{o}}^{-2}(2\omega)} \tag{3}$$

Assuming a fundamental wavelength of $\lambda_0 = 800$ nm and the Sellmeier equations for Barium borate the critical phase matching angle is found to be $\theta = 29.2°$, with this we can set the crystal in such a way to produce SHG.

The Fourier transform has the property that the width of a function in either the time or the frequency domain is inversely proportional to the width in the conjugate space. For a Gaussian pulse, the relation between the time domain width of the measured intensity and width of the pulse in frequency is given by:

$$\sigma_\nu \sigma_t = \frac{2\ln(2)}{\pi} \approx 0.44 \tag{4}$$

where σ_ν and σ_t is the full width half max in the frequency and time domain respectively.

III. METHODS & RESULTS

Now in this experiment we are interested in studying this different set of non linear phenomenon so we use a a spectrometer to detect the different wavelengths being produced after the light interacts with the crystal. The setup implemented is shown in Fig.1 consisting of a half wave plate and a polarizer with which the intensity is modulated .

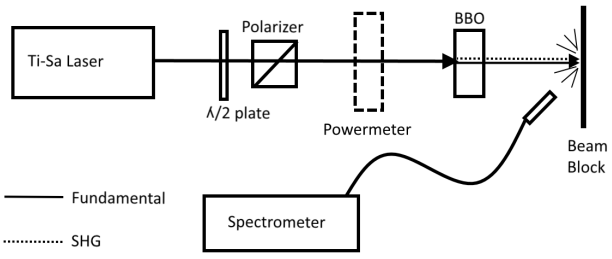


FIG. 1: Set up used to modulate the fundamental beam, detect the SHG and determine the produced signal of the incoming beam interaction with a BBO crystal.

With this we continue to look at the results and analysis of the data collected through the experiment. In this laboratory we are working with the High Intensity TW laser at the Lund Laser Center (LLC). The properties of the laser are listed in Tab. I.

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TABLE I: Properties of the High Intensity TW laser at the Lund Laser Center (LLC).

Average Power	P = 0.6 W
Pulse energy	E = 60 mJ
Pulse repetition rate	f = 10 Hz
Pulse length	$\tau = 45$ fs
Central wavelength	$\lambda = 800$ nm ($\sim T = 2.7fs$)
Laser bandwidth	$\Delta\lambda \approx 30$ nm

The spectrum of the laser source is analyzed with a spectrometer but since the intensity of the beam is too high to measure the beam directly, only the reflected light from a beam block is measured. The spectrum is seen in Fig. 2.

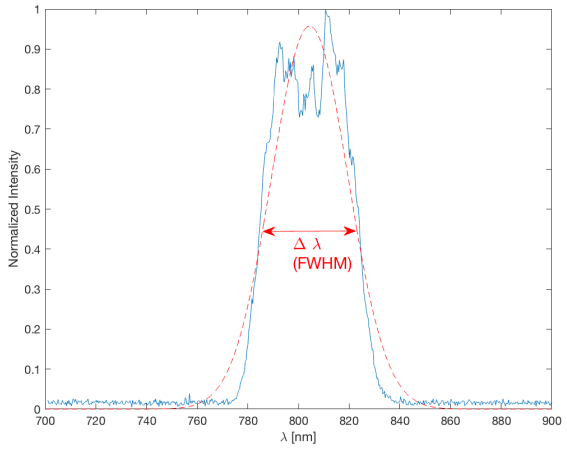


FIG. 2: Spectrum of the fundamental laser frequency measured with a spectrometer. The beam is assumed to be Gaussian and therefore a Gaussian curve is fitted and the FWHM calculated.

The line width (FWHM) of the fundamental is measured to be 35.6 nm at a central wavelength of 804.5 nm and following

$$\tau = \frac{0.44\lambda_0^2}{\Delta\lambda c_0} \tag{5}$$

(see preparation exercise 4), the Fourier limited duration of the pulse is calculated to be 26.7 fs.

A. Spectral properties of the second-order harmonic generation

In the first part of the experiment, we are looking at a second-order harmonic generation (SHG). We are analyzing the length of the frequency doubling crystal on the SHG.

First, we are using a 3mm BBO crystal. As is shown in Ref[1-2] SHG effective conversion efficiency can be written as:

$$d_{eff}(\phi,\theta) = d_{31}\sin\theta + (d_{11}\cos 3\phi - d_{22}\sin 3\phi)\cos\theta, \tag{6}$$

one can tune the angle ϕ by turning the crystal around the beam axis in order to find the highest efficiency (θ given by the cut angle of the crystal, see preparation exercise 2). By changing the incoming angle on the crystal, one can tune the central frequency of the SHG as seen in Fig. 3. This "tunability" is due to the fact that for different incoming angles the phase matching is fulfilled for different central wavelengths. Since the effective conversion efficiency is depending on the angle θ (see Eq. 6), the intensity of the SHG gets worse for central wavelengths further away from 400 nm. Depending on the central wavelength of the SHG one can observe a "hole" in the spectrum of the fundamental at different wavelengths. This is due to the fact the phase matching condition is only fulfilled for a certain part of the fundamental spectrum and therefore the intensity of this part is reduced in the fundamental since these frequencies are converted to the SHG.

For the highest conversion efficiency, a line width of 1.4 nm at a central wavelength of 401.4 nm is measured which corresponds to a Fourier limited pulse duration of 166 fs. This means that the SHG pulse is more than 6 times longer than the fundamental pulse which is to be expected since only a small portion of the fundamental fulfils the phase matching conditions making the bandwidth smaller. We get a longer SH pulse for this same reason since the bandwidth is smaller then the pulse duration is longer as would be expected.

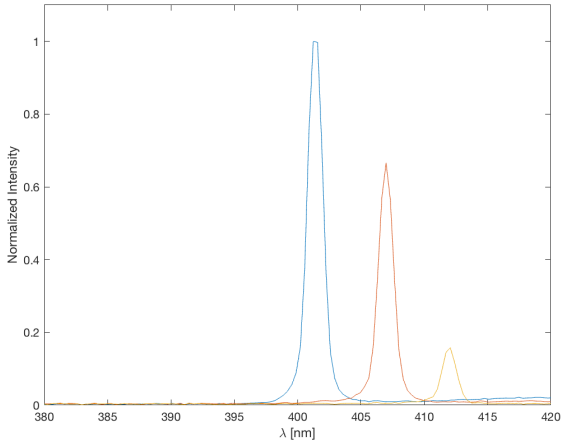


FIG. 3: Spectrum of the SHG for different central frequencies. Central wavelengths at 401.4 nm, 407.0 nm, and 412 nm.

In a second step, we use a 250 μm crystal. The resulting spectrum, as well as the one from the thick

crystal, is seen in Fig. 4. The intensity of the SHG is much higher with the thick crystal, which is due to the fact that in perfect phase matching the intensity is proportional to L^2. The line width at 407.5 nm central wavelength is measured to be 6.7 nm and a Fourier limited pulse duration of 36.0 fs.

The lower pulse duration is due to a smaller temporal walk-off. The temporal walk-off is due to different group refractive indices for different frequencies in dispersive materials. This means that different frequencies have different group velocities and therefore a transform-limited pulse gets wider. If a pulse is already upchirped and the dispersive media has a positive chirp coefficient the pulse gets longer when it travles through the material is, for a negative one it gets shorter or longer depending on the distance traveled in the media, for a downchirped the other way around. In our experiment, the pulse is longer for the thicker crystal.

Since the fundamental pulse was already upchirped the temporal walk-off is stronger for the thicker crystal and the pulse, therefore, gets longer. This effect is stronger for the thicker crystal since the fundamental pulse has more time to interact with the crystal and produce a longer pulse.

With the thin crystal, the central wavelength of the SHG cannot be tuned since the conversion efficiency drops to fast if we do not have the perfect phase matching conditions (efficiency proportional to (length)^2).

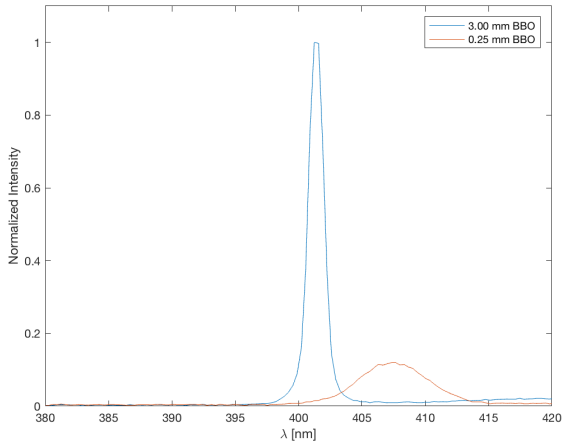


FIG. 4: Spectrum of the SHG for two different thick BBO crystals (3 mm and 250 μm).

Anyhow, as we have seen in preparation exercise 1e) the SHG intensity follows

$$I^{(2)} \sim L^2 \mathrm{sinc}^2\left(\frac{\Delta k L}{2\pi}\right), \tag{7}$$

with sinc(x)=sin(πx)/(πx). Therefore for thinner crystals, the maxima of the intensity are closer to each other. This means that we have to turn the crystal by a smaller angle to get to the next maxima. We have a global maximum at $\Delta k = 0$ and the next maxima at $\frac{\Delta k}{2} = \frac{3\pi}{2}, \frac{5\pi}{2}$, etc. These side maxima have a relative intensity of 1/22 or 1/63 respectively compared to the main maximum. In Fig. 6 the first main fringe is seen, which is measured by changing the incoming angle of the crystal. These maxima are called Marker's fringes and there intensity drops quickly for higher orders as dicussed before for this we can only observe the first order. The measured relative intensities of the first side maxima are 1/16 and 1/40. These results are in the same order of the calculated theoretical values but since the adjustment of the angles is only done by hand the values are not reliable.

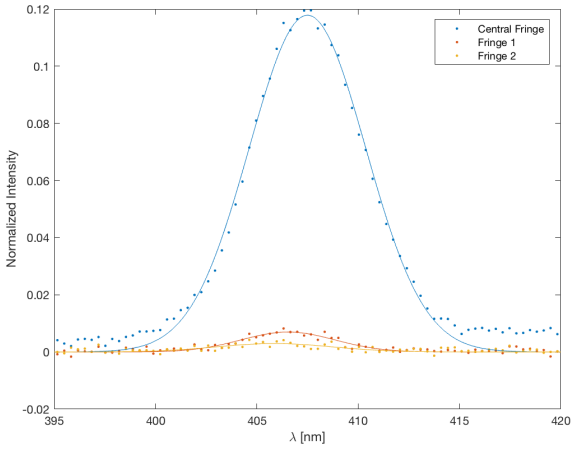


FIG. 5: Spectrum of the SHG for Marker's fringe recorder at different times. We can only observe one fringe since the intensity drops the fringe order.

The simulations of the SHG intensity following Eq. 7 for the two crystals are seen in Fig. 6. It is seen that for the thick crystal the Marker's fringes are much closer to each other and for the thin crystal the central peaks are much wider.

B. Intensity dependence of the second harmonic

In order to modulate the intensity of the laser beam, an attenuator is used. Since the beam from the laser is linear polarized one can place a half-wave plate and a polarizer. In this set up the full power of the beam passes the polarizer. If one now turns the half-wave plate, the polarization direction changes and only part of the beam passes the polarizer. Therefore one can tune the power of the beam.

This set up is used to analyze the intensity dependence

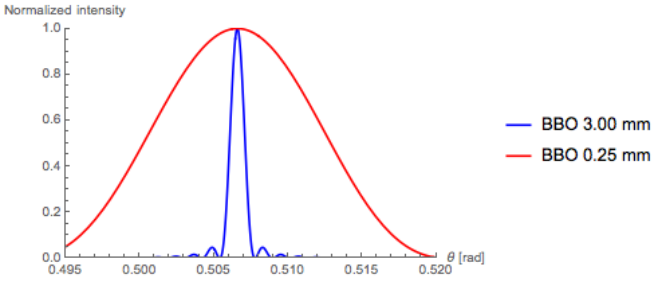


FIG. 6: Simultation of the intesity of the SHG for two different thick BBO crystals. The intensity is normalized to the main fringe and the angle θ describes the incoming angle relative to the optical axis.

of the SHG. The intensity of the fundamental and the SHG are measured for different half-wave plate angles. When the fundamental interacts with the BBO crystal it gets partly depleted by the SHG and the intensity of the fundamental is much stronger then the SH signal its not practical to measure with a powermeter. It's worth mentioning that the fundamental's intensity is controlled independently before the crystal.

In Fig. 7 the intensity measurements are shown in a logarithmic plot with a fit. The calculated intensity relation is $I_{SH} \sim I^{2.4}$. Since this is the second harmonic generation we expected a result of the exponential close to 2.

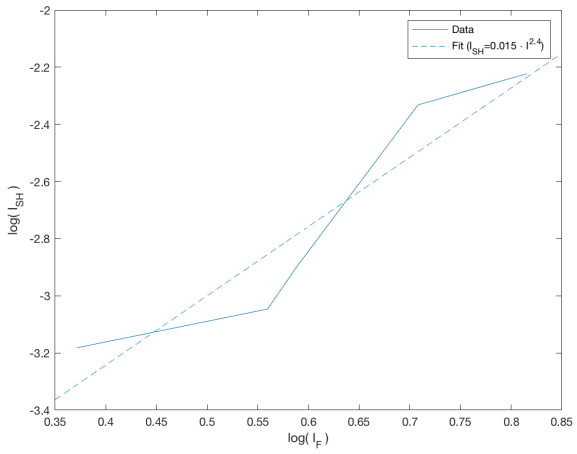


FIG. 7: Logarithmic plot of the SHG intensity for different fundamental intensities with a linear fit.

C. Generating a third-order harmonic in air

In this part of the experiment, we generate a third-order harmonic (TH) in air. In order to do this, we need a higher intensity of the beam achieved by focusing the beam. A set of two lenses is used to collimate the beam a plano-convex lens with focal length 40 mm focuses the beam and a 100 mm lens defocus it. The distance between the lenses is the sum of their focal lengths and on general the second lens has a smaller focal length in which produces a collimated beam with a smaller beam diameter.

In order to measure only the TH frequency, we separate it from the fundamental with a Pellin-Broca prism. If we let the outgoing beam propagate on a white paper the TH gets visible. This is due to the fact that white paper is often treated with optical brightener, which absorbs UV light and emits blue light. Since the TH is at 267 nm it is UV and gets visible. The measured spectrum is seen in Fig. 8

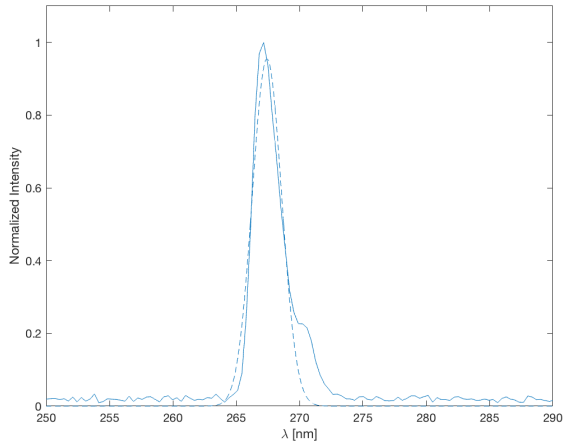


FIG. 8: Spectrum of the third-order harmonic generation at 267nm.

We can not see a SHG since air is a gas and therefore centrosymmetric. Centrosymmetric materials have the property that their electric potential has the symmetry V($\vec{r}$)=V(-$\vec{r}$) and therefore the polarization is always parallel to the electric field. This means that there is no E^{2n} ($n \in$) term in the polarization. Therefore the harmonic generation with the highest intensity is the third order.

IV. CONCLUSIONS

The most note worthy results of this experiment were the observed SHG of the thick crystal had a smaller bandwidth than that of the thin one. Phase-matching in the thick BBO is given for a much narrower frequency range, give of by the effect of the materials large dispersion that in part id given by the different path lengths that each photon pair must travel. We also conclude that we don't observe SHG in air since its a centrosymetric material. Due to this we can study its third order effects which were observed in the experiment with a low intensity since

there is such a low probability of three photon absorption in the material.

[1] D. Meschede, ed.Optics, Light and Lasers: the practical approach to modern aspects ofphotonics and laser physics. 1. reprint. Weinheim: Wiley-VCH, 2005, IX, 410 pages.isbn: 3-527-40364-7.
[2]Masada, G. (2011). Efficient Generation of Second Harmonic Wave with Periodically Poled MgO:LiNbO3. Tamagawa University Quantum ICT Research Institute Bulletin, 1(25_28 (2011) .
[3] M. N. Polyanskiy.Refractive index database.https://refractiveindex .info.Accessed on 2019-06-10.
[4]R. A. Ganeev et al. "Third-harmonic generation in air by use of femtosecond radiation intight-focusing conditions". In:Appl. Opt.45.4 (Feb. 2006), pp. 748–755.doi:10.1364/AO.45.000748.url :http://ao.osa.org/abstract.cfm?URI =ao-45-4-748.AlbinHedse, MariusPlach, Rasmuslyngbye pedersen12.