$\underline{Nonlinear\ Ultrafast\ Optics}$
$\underline{supplementary\ material}$

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Supplementary section 1: Additional Theoretical Background.

1.1 polarization vector as electric field source.

If we look at the time varying polarization, and applying it to the wave equation in nonlinear media we get:

$$(1)\ \nabla^2\widetilde{\boldsymbol{E}} - \frac{n^2}{c^2}\frac{\partial^2\widetilde{\boldsymbol{E}}}{\partial t^2} - \frac{1}{\epsilon_0 c^2}\frac{\partial^2\widetilde{\boldsymbol{P}}^{NL}}{\partial t^2} = 0$$

Where n is the linear refractive index and c is the speed of light in vacuum. The $\frac{\partial^2\widetilde{\boldsymbol{P}}^{NL}}{\partial t^2}$ represent the acceleration of the charges in the medium, and the theory of electromagnetism tells us that, accelerating charges generate electromagnetic radiation that in this case act as a source of radiation [1].

Our experiment is about SHG, therefore we will discuss the second term in the infinite polarization series: $(2)\ \widetilde{\boldsymbol{P}}^{(2)} = \epsilon_0\chi^{(2)}\widetilde{\boldsymbol{E}}^2$

We can assume that the incident electric field time dependency is in a form of:

$$(3)\ \widetilde{\boldsymbol{E}}(\boldsymbol{r},t) = \boldsymbol{E_0}e^{-i\omega_{FF}t}$$

when we place eq.3 in eq.2 and then in eq.1 we get $\widetilde{\boldsymbol{E}}(r,t)$ solution that have terms with $2\cdot\omega_{FF}$ time dependency [1]. These second harmonic solutions, testifies the expected doubled frequency spectrum (SHG) that we try to observe in our experiment.

1.2 Phase matching

The condition of phase matching can be described physically in a classical way, such that the generated **E** waves that the vibrating dipoles creates, when the incident laser passes through the crystal, has the same phase, and creates constructing interference when the beam come out of the crystal [1]. Therefore, the condition that we want to achieve is:

$$(4)\ \Delta k = k_1 + k_2 - k_3 = 0$$

This condition simply translates to a condition on the refractive indexes of the beams in the media:

$$(5)\quad \frac{n_1\omega_1}{c} + \frac{n_2\omega_2}{c} = \frac{n_3\omega_3}{c}$$

When n_1 is refraction index of beam one, n_2 is refraction index of beam two and n_3 is refraction index of the output beam. In our experiment the incident light frequencies ω_1 and ω_2 are equal (the source is monochromatic coherent laser). In addition, the output frequency ω_3 that we seek is twice the fundamental frequency of the laser. And the final condition is:

$$(6)\ n(\omega) = n(2\omega)$$

This condition is supposedly not possible to achieve because that the refraction index mostly is a monotonic function of ω . To overcome this, we used birefringent crystals. Birefringence is optical property of a material having refractive index depends on the polarization and direction of light. Not all crystals display birefringence, it depends on their inner symmetrical built, and if they cannot it can be said that they are not phase matchable. In our experiment we used negative uniaxial crystal, which preforms the simplest type of birefringence [2]. Uniaxial means that there is a single optical axis direction - $\hat{c}$ that characterizes the crystal. Light polarized perpendicular to the plain containing the propagation vector $\boldsymbol{k}$ and optical axis $\hat{c}$, is called ordinary polarization. This light experience refractive index that depends only on its frequency and is called ordinary index - $n_o(\omega)$. The other direction of polarization is when the light polarized in the plane that contains $\boldsymbol{k}$ and $\hat{c}$. This type of polarization is called extraordinary polarization and experiences refractive index that depend on the angle θ between $\boldsymbol{k}$ and $\hat{c}$ - $n_e(\theta)$ [1].

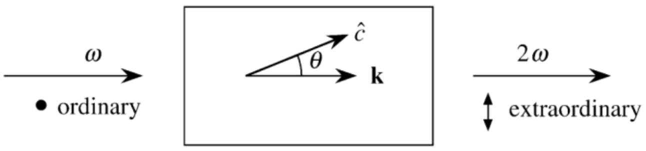

Figure (1)- ordinary and extraordinary light waves orientation.

The relation between n_e and θ derived in couple of articles for example Born and Wolf (1975, Section 14.3) [1]:

$$(7)\ \frac{1}{n_e(\theta)^2} = \frac{\sin^2\theta}{\bar{n}_e^2(2\omega)} + \frac{\cos^2\theta}{n_0^2(2\omega)}$$

In this formula, $\bar{n}_e$ is the principal value of the extraordinary refractive index.

Another useful relation derived in Boyd's book [1] is the exact relation between SHG intensity as a function of input beam and crystal properties:

$$(8)\ I_{SH} = \frac{8d_{eff}\omega_{SHG}^2 I_{FF}^2}{n_0^2(\omega_{FF})n_e(\omega_{SHG})\epsilon_0 c^2} L^2 sinc^2\left(\frac{\Delta kL}{2}\right)$$

Where ω_{SHG} is the second harmonic frequency, I_{FF} is the input beam intensity, $n_{e/o}$ are the extraordinary/ordinary refractive indexes of the crystal, L is the effective length that the beam passes through the crystal, Δk is phase mismatch, ϵ_0 is the vacuum permittivity constant, c – speed of light, and $d_{eff} = \frac{1}{2}\chi^{(2)}$ is half of second order susceptibility constant in the crystal.

Supplementary section 2: Detailed Experimental Setup

The experiment conducted on optical table, that on top of it were placed the following optical components:

- MIRA900 – ultrafast femtosecond laser. Labeled as class 4 laser that can emit extremely powerful radiation. The laser has capability of mode locking, which means that it is able to produce shot pulses in time and wide (about 10 nm) in spectrum. We used the laser while it was in mode-locking. [3].
- BBO crystal. negative uniaxial ($n_e < n_o$). Designed such that for direct perpendicular incident angle, the SHG is maximal.
- 2 focal lenses- an optical setup of two aligned lenses, meant to focus the beam.
- Thorlabs continuous ND (neutral – density) wheel filter. This filter reduces intensity of all wavelengths equally, allow us to control beam's intensity by spinning the wheel [4].
- Thermal mobile power meter- measures the beam power after the ND filter. Has a thermal sensor that converts temperature difference to voltage then to incident beam's energy [5].
- Spectrometer connected to optic fiber – measures the spectrum, which means counts of incident photons per wavelength per specific time. The spectrometer interacts with "Oceansview" software to present real time data [6].
- Optical mirrors – directed the beam to its selected optical path.

Other optical components were placed but their details are written in the main paper.

Supplementary section 3: Gaussian fit for the fundamental frequency (FF) and for the SHG.

To determine the wavelengths of the fundamental frequency (FF) and second harmonic generation (SHG), Gaussian fits were performed on the data acquired from the spectrometer using MATLAB. For the FF five burst measurements (see section 5) were conducted with the same integration time, and for SHG ten burst measurement were done with much larger integration time. The resulting data collected from the burst measurements was averaged. The wavelength error was treated as a uniform distribution error, while the intensity (counts) error was considered a statistical error. For the FF, a Gaussian fit was applied using the following equation:

$$(9)\ y = a_0 \cdot e^{-\frac{(x-a_1)^2}{a_2^2}} + a_3$$

Where x stands for wavelength, y as intensity, a_0 denotes the normalization factor, a_1 represents the FF wavelength, a_2 denotes the fit uncertainty, and a_3 accounts for systematic errors such as averaged background noise.

The fit and residuals that we for fundamental wavelength:

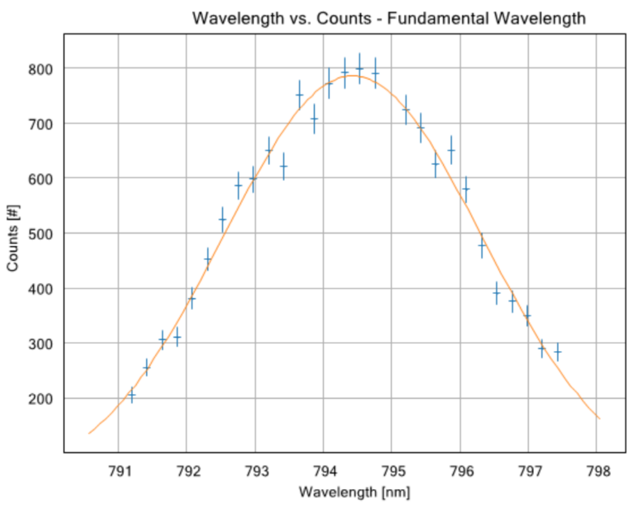

Figure (2) - gaussian fitting for fundamental laser wavelength.

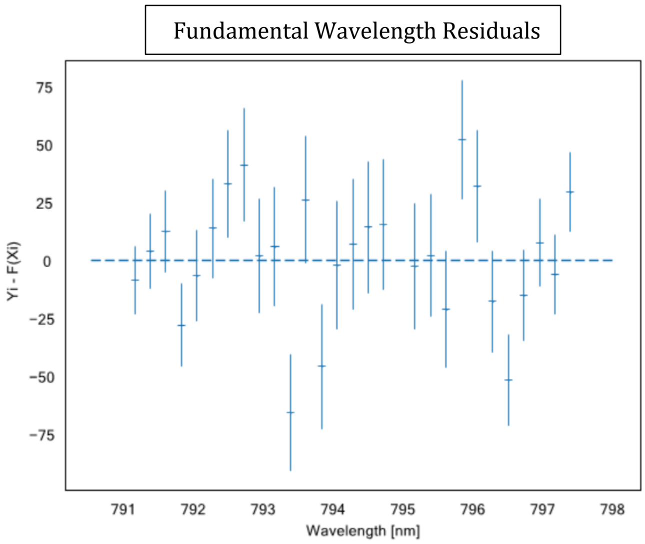

Figure (3) - residuals for fundamental laser wavelength.

Parameter	Value	Relative error
χ^2_{red}	1.3	-
p_{val}	0.13	-
a_0	770 ± 65	8.4%
a_1 [nm]	794.419 ± 0.028	0.0035%
a_2 [nm]	1.91 ± 0.15	8.1%
a_3	25 ± 69	270%

Table (1) – fitting parameters for fundamental wavelength.

The fit quality parameters are in the desired range. that can indicate that indeed the fitting function is a good fit for data. The residuals are showing random scattering around the curve. It can be said that the fit is good overall.

The resulted fundamental wavelength:

$$\lambda_{FF} = 794.419 \pm 0.028 \text{ [nm]}$$

And for second harmonic wavelength:

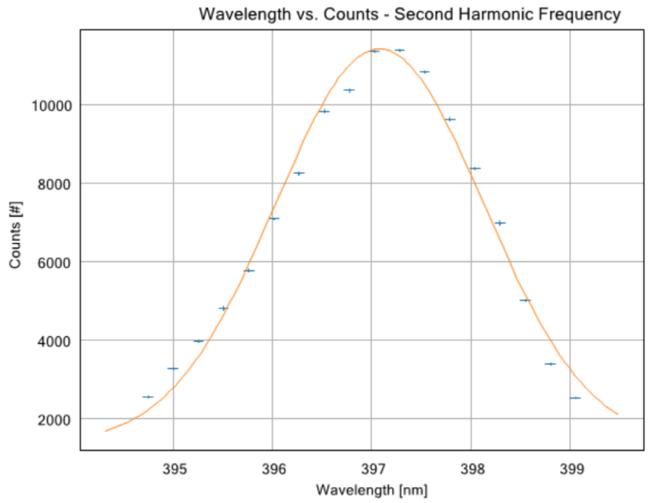

Figure (4) – Second Harmonic spectrum.

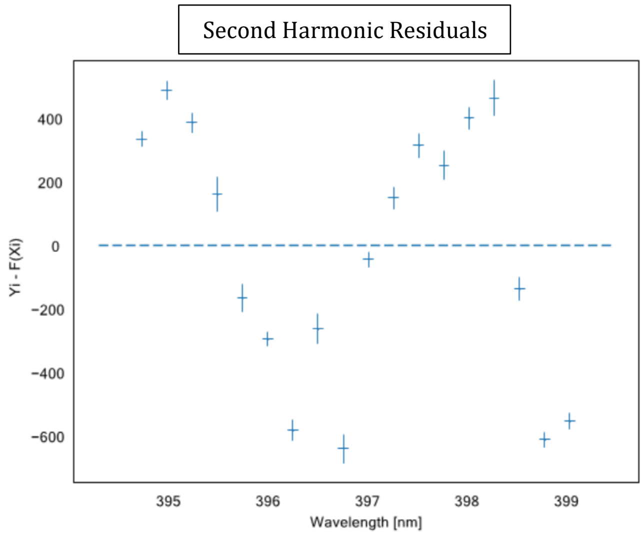

Figure (5) - residuals for Second Harmonic Spectrum.

Parameter	Value	Relative error
χ^2_{red}	6.1	-
p_{val}	0	-
a_0	10040 ± 540	5.3%
a_1 [nm]	397.080 ± 0.030	0.0076%
a_2 [nm]	1.050 ± 0.074	7.1%
a_3	1380 ± 550	40%

Table (2) – fitting parameters for second harmonic spectrum.

The fit quality parameters are not in the desired range. However, the graph shape looks like it fits the data. The out-of-range fit parameters may also be caused by small errors in y axis as can be showed in the residuals. The errors are small because the statistical error in counts is less significant when the counts are high.

The resulted fundamental wavelength:

$$\lambda_{SHG} = 397.080 \pm 0.030 \text{ [nm]}$$

Supplementary section 4: Numerical Simulation $I_{SHG}(\alpha)$.

In this section a numerical simulation was done to analyze the connection between the incident angle of the laser - α to the SHG intensity, based on eq. 8. We conducted the simulation on couple of different L_0 values that represent the depth of the crystal.

First, we need to consider that when the incident beam passes from the air medium to the crystal medium, we need to apply Snell's law to find the angle θ' which is the direction of $\hat{k}$. By manufacturing details of the crystal, we know that when $\alpha = 0$ (no refraction), we get maximum SHG intensity for laser beam of 800 [nm]. Combining that knowledge and applying the condition in eq. 6 to the theoretical formula in eq.7, we can find the angle which we get the desired phase matching:

$$(9)\ \theta_0 = \sin^{-1}\left[\left(\frac{\frac{1}{n_0(\omega)^2} - \frac{1}{n_0(2\omega)}}{\frac{1}{n_e(2\omega)^2} - \frac{1}{n_0(2\omega)^2}}\right)^{-\frac{1}{2}}\right]$$

And in total the angle θ between $\hat{k}$ and $\hat{c}$ is:

$$(10)\ \theta(\alpha) = \theta_0 + \sin^{-1}\left(\frac{\sin(\alpha)}{n_0(\omega)}\right)$$

Second, we found L by simple geometrical condition:

$$(11)\ L = \frac{L_0}{\cos(\theta')} = \frac{L_0}{\cos\left(\sin^{-1}\left(\frac{\sin(\alpha)}{n_0(\omega)}\right)\right)} = \frac{L_0}{\sqrt{1 - \frac{\sin^2(\alpha)}{n_0^2(\omega)}}}$$

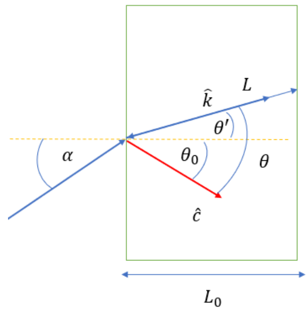

Figure (2)- Illustration of the geometrical path of the beam inside the crystal.

Third, we combined all the developed values to eq. 8 and got the fully developed connection:

$$(12)\ I_{SH}(\alpha) \propto \frac{L_0^2}{1 - \frac{\sin^2(\alpha)}{n_0^2(\omega)}} sinc^2\left(\frac{2\pi L_0}{\lambda_{FF}\sqrt{1 - \frac{\sin^2(\alpha)}{n_0^2(\omega)}}}\left(n_0(\omega) - \left(\frac{\sin^2\left(\theta_0 + \sin^{-1}\left(\frac{\sin(\alpha)}{n_0(\omega)}\right)\right)}{\bar{n}_e^2(2\omega)} + \frac{\cos^2\left(\theta_0 + \sin^{-1}\left(\frac{\sin(\alpha)}{n_0(\omega)}\right)\right)}{n_o^2(2\omega)}\right)^{-\frac{1}{2}}\right)\right)$$

While the known constants we used are [7]:

$n_0(2\omega) = 1.6934$, $\bar{n}_e(2\omega) = 1.5687$

$n_o(\omega) = 1.6614$

And the calculated values:

$\theta_0 = 29.01°$ calculated from eq. 9.

$\lambda_{FF} = 794.419\ [nm]$ extracted from section 3.

Then defined a range of α and plotted normalized graphs of intensity vs. α.

Supplementary section 5: MATLAB Code Specifications.

The measurements were done by dedicated MATLAB codes, specifically written to automate the measuring process. The code has some important user changeable parameters: **Integration time**, is the time that the spectrometer sensor collects the data per single measure. **Burst number** is the number of times that a measure is taken. **Jump** is the angle difference that the motor moves each step.

The code has also functionality to rotate the stage do desired angle or to continuously move stage by user interface.

Supplementary section 6: Sinc squared fitting and error analysis.

The goal of this part was to fit sinc squared fitting of I_{SHG} vs. incident angle α. The fit relied on gaussian fits for each measured angle according to eq. 9.

From each fitting we extracted a_1 – the peak wavelength, a_2 the standard deviation of the peak, and a_0 peak intensity (counts). Assuming the SHG intensity has gaussian distribution we calculated it by the following formula:

$$(13)\ I_{SHG} = \int_{-\infty}^{\infty} a_0 e^{-\frac{1}{a_2^2}(x-a_1)^2} = a_0 a_2 \sqrt{\pi}$$

The error of I_{SHG} was the statistical error of the closest point to the maximum of the fitting. Meaning the intensity of the wavelength closest to a_1.

The error of α was resolution error obtained by the difference within two jumps, in our case 0.05°.

Supplementary section 7: Sinc Squared without secondary peaks fitting results.

We conducted another fitting dismissing the edges of the data, trying to get better results.

The fits and residuals that received:

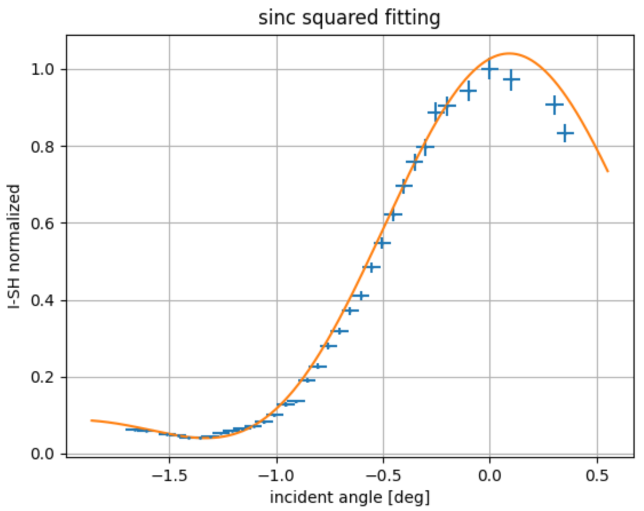

Figure (6) - $I_{SHG-normalized}(\alpha)$ without edges.

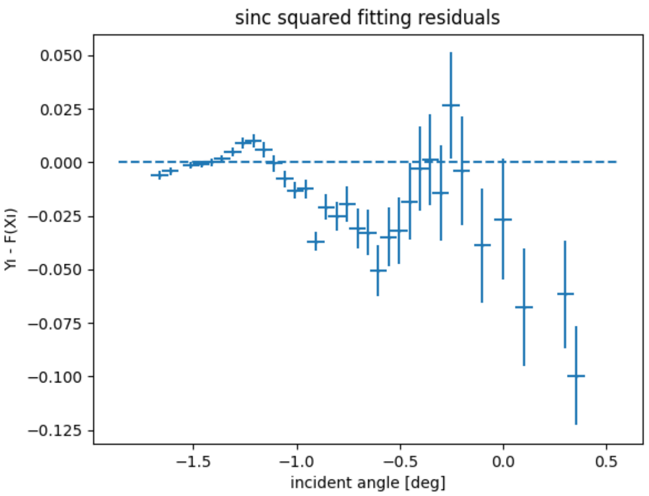

Figure (7) - $I_{SHG-normalized}(\alpha)$ without edges residuals.

Parameter	Value	Relative error
χ^2_{red}	1	-
p_{val}	0.4	-
a_0 [m]	$2.388 \cdot 10^{-4} \pm 4.1 \cdot 10^{-6}$	1.7%
a_1 [deg]	$-1.41 \cdot 10^{-3} \pm 3.1 \cdot 10^{-4}$	21%
a_2	0.0402 ± 0.0014	3.6%

Fit quality parameters are in the desired range. The fit seems to match the data. We can consited the fit as a success.

The extracted L_0 value from the fitting:

$$L_0 = 2.388 \cdot 10^{-4} \pm 4.1 \cdot 10^{-6}\ [m]$$

5) References

[1] Boyd, Robert W. *Nonlinear optics*. Academic press, 1992. Pages 1-10, 69-84.

[2] Wikipedia. Birefringence url: https://en.wikipedia.org/wiki/Birefringence

[3] Wikipedia. Mode - Locking url: https://en.wikipedia.org/wiki/Mode_locking

[4] Wikipedia. Neutral – density filter url: https://en.wikipedia.org/wiki/Neutral-density_filter

[5] Thorlabs power meter and sensor tutorial: https://www.thorlabs.com/newgrouppage9.cfm?objectgroup_id=6188#:~:text=In%20a%20thermal%20power%20sensor,the%20radiator%20of%20the%20sensor.

[6] Nonlinear optics instructions from Moodle Lab C course website, written by Eyal Bahar, 2020.

[7] Refractive index database url: https://refractiveindex.info/