Nonlinear Optics

Laboratory exercise in
Advanced Optics and Lasers VT 2015

Understanding the non-linear theory of light.

Chen Guo
Atomic Physics Division
Lund University
chen.guo@fysik.lth.se
Introduction

The aim of this laboratory exercise is to investigate nonlinear optical effects that arise when an intense laser beam interacts with matter. In the first part of the laboratory exercise, we will convert an invisible infrared laser beam into a visible blue laser beam using a second-order optical process in a phase-matched doubling crystal. Macroscopic phase-matching of the process in the entire crystal is important for an efficient photon conversion since the microscopic non-linear interaction is small. We will experimentally study how the nonlinear conversion depends on the length of the crystal. Depending on the application, you will learn how to choose the best length for the doubling crystal so that either the conversion energy is maximized or so that the length of the generated pulse is short.

We will then demonstrate that nonlinear effects also appear in less complicated situations if the laser intensity is high: Focusing the intense laser beam in air results in a third-order optical process that converts the infrared laser into an ultraviolet laser beam.

Nonlinear optics presents useful applications in spectroscopy. Nonlinear laser interaction provides the possibility of generating laser beams with high photon energy from high power lasers with low photon energy. This is useful because it is difficult to build high photon energy lasers. Frequency doubling and tripling are the trademarks of nonlinear optics, but there are also other useful phenomena. Self-phase modulation is a third order effect which can be used to broaden the spectrum of a laser pulse. In ultra-fast optics scientists are using such spectrally broadened pulses to create laser pulses with duration of a few femtoseconds. This corresponds to a laser pulse with a single electric field oscillation!
Consider the following questions:

1) In this exercise we will derive the nonlinear interactions for the second harmonic using the scalar wave equation for the electric field (Eqn. 21.1-3):

\[ \nabla^2 \widetilde{E} - \frac{n^2}{c^2} \frac{\partial^2 \widetilde{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \widetilde{P}}{\partial t^2}, \]

where \( \widetilde{E} = \widetilde{E}(t, \vec{r}) \) is the electric field, \( \widetilde{P} = \widetilde{P}(t, \vec{r}) \) is the polarization. \( n = n(\omega) \), \( c \) and \( \mu_0 \) are the refractive index, vacuum speed of light and magnetic permeability. Mathematically, we can regard the wave equation as an inhomogeneous partial differential equation where the electric field wave is driven by the polarization of the media. The approximation we use to solve this difficult problem is called the First Born approximation.

We model the incoming laser field as a plane wave of constant strength:

\[ \widetilde{E}_1(t, \vec{r}) = 2E_1 \cos(k_1 z - \omega_1 t) = E_1 \left( e^{-i(k_1 z - \omega_1 t)} + e^{i(k_1 z - \omega_1 t)} \right), \]

where \( k_1 \) is the wavevector and \( k_1 = n_1 2\pi / \lambda_1 = n_1 \omega_1 / c \) with \( \lambda_1 \) being the vacuum wavelength and \( n_1 = n(\omega_1) \); and \( E_1 \) is a real constant.

a) We assume that the polarization is instantaneously induced by the laser:

\[ \widetilde{P}(t, \vec{r}) = \varepsilon_0 (\chi^{(1)} \widetilde{E}_1 + \chi^{(2)} \widetilde{E}_1^2 + \chi^{(3)} \widetilde{E}_1^3 + ...) = \widetilde{P}^{(1)} + \widetilde{P}^{(2)} + \widetilde{P}^{(3)} + ... \]

The second order polarization

\[ \widetilde{P}^{(2)} = \varepsilon_0 \chi^{(2)} \widetilde{E}_1^2 = \varepsilon_0 \chi^{(2)} E_1^2 \left( e^{-i(2k_1 z - 2\omega_1 t)} + e^{i(2k_1 z - 2\omega_1 t)} + 2 \right) \]

- Now calculate the acceleration of the polarization (its second time derivative) which is relevant for the wave equation, use a complex exponentials formula. 

Hint: The acceleration of the polarization should be complex exponentials oscillating at \( \pm 2\omega_1 \). These are the source for the second harmonic field!

b) To find the newly generated electric field, we assume it to be a complex plane wave that can vary in strength as it propagates along the \( z \) axis:

\[ \widetilde{E}_2(t, \vec{r}) = E_2(z) \exp(ik_2 z - i2\omega_1 t). \]

- Calculate the corresponding Laplacian:

\[ \nabla^2 \widetilde{E}_2 = (\partial^2 / \partial x^2 + \partial^2 / \partial y^2 + \partial^2 / \partial z^2) \widetilde{E}_2. \]

Since the new field will grow very slowly, \( \frac{\partial^2 E_2}{\partial z^2} \ll \left| \frac{i2k_2 \partial E_2}{\partial z} \right| \) you may neglect the term containing the second derivative of \( z \).

c) Verify that the total left hand side of the wave equation becomes:

\[ \nabla^2 \widetilde{E}_2 - \frac{n_1^2}{c^2} \frac{\partial^2 \widetilde{E}_2}{\partial t^2} \approx i2k_2 \frac{\partial E_2}{\partial z} \exp(ik_2 z - i2\omega_1 t). \]
d) Solve the approximate wave equation for $E_z(z)$ for one of the complex polarization source terms:

$$i2k_z \frac{\partial E_z}{\partial z} \exp(ik_z z - i2\omega t) = -4\omega_l^2 \varepsilon_0 \mu_0 \chi^{(2)} \exp(2k_z z - i2\omega t).$$

- Assume that the material starts at $z = 0$ and ends at $z = L$. This implies the following boundary condition: $E_z(z) = 0, \ z < 0$. Write your answer using the wavevector mismatch, $\Delta k = 2k_1 - k_2 = 2\omega_l (n_1 - n_2) / c$.

*Hint*: This is a first order differential equation which you can solve by simply integrating the right hand side over $z$.

e) Calculate an intensity of the second harmonic light after passing the material?

*Hint*: The intensity is given by the time average of the square of the electric field,

$$I(z) = \varepsilon_0 c \left< \tilde{E}_z^2(z) \right> = \frac{\varepsilon_0 c |2E_z(z)|^2}{2} \text{ in vacuum for } z > L \text{ and } e^{-iz} + e^{iz} = 2 \cos z.

**General comment:**
In this problem we have not considered the polarizations of the beams. For second harmonic generation we have perpendicular polarizations, $\tilde{E}_1 \perp \tilde{E}_2$. You can use the same approach to get a similar expression for the third harmonic, oscillating at $3\omega_l$!

2) Explain how the second-harmonic conversion can be maximized using birefringent crystals, *i.e.* how phase-matching can be achieved ($\Delta k = 0$).

Numerically determine a BBO cut-angle for which second harmonic generation from laser source that will be used in the laboratory exercise is phase-matched, use Type I phase-matching (the fundamental light propagates as ordinary wave and second harmonic as extraordinary).

*Hints:*
- The refractive index, $n_x$, of a crystal depends on the wavelength of the light, $\lambda$, and can be calculated using the Sellmeier equations (see equipment section) where “x” is either “o” for the ordinary- or “e” for the extraordinary axis.
- Use the Ellipse equation for birefringent crystals:

$$\frac{1}{n_e(\theta)^2} = \frac{\sin^2 \theta}{n_o^2} + \frac{\cos^2 \theta}{n_o^2}.$$

3) Calculate a phase matching bandwidth (look equation 21.2-30 from *Fundamental of Photonics*, B. E. A. Saleh, second edition) of a 3 mm long Beta Barium Borate (BBO) crystal cut for second harmonic generation (Type I – ooe) of 800 nm radiation. Remember to use group refractive indexes.

4) Calculate transform limited pulse duration of a pulses with spectral bandwidth of 30 nm centered at 800 nm and a pulses with spectral bandwidth of 15 nm centered at 400 nm. Assume a Gaussian pulse shape.

*Please read though the laboratory instructions.*
Practical laboratory exercises

Beam time at the High Intensity TW laser, 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 ($\sim T = 2.7$ fs)
- Laser bandwidth $\Delta\lambda \approx 30$ nm

Beta Barium Borate – BBO

- Thin BBO $L = 0.25$ mm
- Thick BBO $L = 3$ mm

Refractive index of BBO (*Sellmeier equation*)

\[ n_x = \sqrt{A_x + \frac{B_x}{\lambda^2} - D_x \lambda^2}, \quad [\lambda \text{ in } \mu\text{m}] \]

<table>
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<tr>
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<th>$A_x$</th>
<th>$B_x$</th>
<th>$C_x$</th>
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Optics:

- Attenuator (Half-wave plate controlled by motorized rotation stage and polarizer),
- Planar-convex lenses ($f_1 = +100$ mm, $f_2 = +50$ mm),
- Pellin-Broca prism.

Measuring devices:

- Avantes spectrometer with optical fiber
- Power meter
**Laser safety**

The laser system that we are using in this lab is a pulsed high energy laser (class 4). **Use protective goggles at all times.** Remove watches and rings! Stand up in the lab at all times. The beam is invisible to our eyes and it can only be seen though the use of fluorescence from yellow cards. It is important that you never insert or exit any optical component from the beam line before blocking the beam with a black metal wall. Always block the beam before changing the setup. Install one component at the time and carefully follow the beam using the yellow cards. Never let the beam propagate freely in the room.

1. **Fundamental laser frequency**

**Spectral properties of the mode-locked laser:**

In this laboratory exercise we are using laser pulses that are generated and amplified using a Titanium-Sapphire stretched-pulse amplifier (Chirp Pulse Amplification method). The laser has a mode-locked oscillator which results in a large spectral bandwidth and a pulsed output. The maximal effective intensity of the pulses from this laser system is extremely high. In fact, this laser system is currently used for studies of high-order harmonic generation (HHG), which is a nonlinear optical process beyond the perturbative regime. We will limit the effective intensity of the pulses by aperturing the beam and by not focusing it very tightly, so that the optical non-linear processes remain in the perturbative regime.

![Diagram](attachment:image.png)

**1.1 Spectral output of the laser system:**

a) Let the beam propagate onto a beam block.

b) Aim the spectrometer’s fiber with “scattering block” into the light. Explain the observed spectra.

c) **Save the spectra.** Estimate the Fourier limited duration of the laser pulses assuming a Gaussian spectral shape.
2. Second-order harmonic generation

2.1 Spectral properties of different doubling crystals:

We now want to study the second-order emissions for a few different doubling crystals. The second-order polarization is proportional to the square of the incident electric field: $P^{(2)} \sim \chi^{(2)}E^2(t)$. The nonlinear constant, $\chi^{(2)}$, is a material constant. In particular we want to see how the length of the doubling crystal affects the emission. We use two different BBO crystals (see equipment) and study the spectral properties using a spectrometer with a range around 400 nm.

2.1.1 Thick doubling crystal:

a) Let the beam propagate onto a beam block.
b) Aim the 800 nm spectrometer fiber with scattering block into the light.
c) Insert the thick BBO (3 mm) into the beam. Carefully adjust the crystal angle to maximize the blue light.
d) **Save the spectra.**
e) Estimate the Fourier limited duration of the generated pulse and compare with the fundamental pulse duration.
f) Explain the observed “tunability” range. **Save the spectra** for different central wavelength.
g) Identify and measure the Maker’s fringes.
h) Study the IR spectra from the “scattering block”. Explain the origin of the “hole” in the spectra.

2.1.2 Thin doubling crystal:

a) Change for the thin BBO (250 μm).
b) **Save the spectra.**
   Estimate the Fourier limited duration of the generated pulse and compare with the fundamental pulse duration.
c) What is a temporal walk-off? (Page 989)
d) Explain why the pulse is shorter than for the thick crystal.
e) Can the central frequency be tuned?
f) Is the bandwidth of the second-harmonic limited by the bandwidth of the fundamental field or by phase-matching? Estimate how broad the ideal spectra of the second-harmonic would be from the fundamental spectra.

**ASSIGNMENT I:**

In this assignment you will write a program in *MATLAB* that simulates the experimental results for the **thick** crystal, L= 3 mm.

Use the optimal cut-angles, as found in preparatory exercise 2). Then use the Sellmeier equations for BBO to calculate the refractive indices: \( n_o(\lambda) \) and \( n_e(\lambda) \). Calculate the extraordinary index for the phase-matched doubled frequency, \( n_e(\theta,\lambda/2) \), using the Ellipse equation. Use the result of preparatory exercise 1) as the conversion factor for doubling:

\[
I^{(2)}(\lambda/2) \sim \sin^2(\Delta kL/2) / \Delta k^2 \quad \text{and} \quad \Delta k = 4\pi \left[ n_e(\theta,\lambda/2) - n_o(\lambda) \right] / \lambda.
\]

The absolute value of the conversion is not required. Compare the calculated crystal length with the experimental results. How do the width of the central peak compare? How does the spacing of the Maker fringes compare?

**2.2 Intensity dependence of the second harmonic**

We will now investigate how the second-order conversion depends on the energy of the fundamental pulses. The energy of the incident laser pulses is controlled using a half-wave plate (\( \lambda/2 \) plate) and a polarizer.

**2.2.1 Adjusting the beam power:**

a) Explain how a half-wave plate and a polarizer can be used to build an attenuator that reduces the power of a linearly polarized beam.  
*Hint:* Transmission of p-polarized light is 100% at Brewser's angle.

b) Make a series of measurements of the second harmonic power as a function of the fundamental power. What is the problem of measuring the power of the infrared beam after the generation? Is it better to use the thin or the thick crystal? Discuss how to conduct the experiment and how to carry out the measurements.
c) Analyze the data from 2.2.1 b). Present the data with \( \log(I_1) \) on the x-axis and \( \log(I_2) \) on the y-axis. Determine the numerical value for the exponential. Is the experimental data indicating that the process we study is of second-order? 
*Hint:* Use \( \log(I') = x \log(I) \) to determine the exponent, \( x \), using a linear fit.

3. **Third order harmonic generation**

3.1 **Generating a third-order harmonic in air:**

In this part of the laboratory exercise we will use air to generate a third harmonic of the fundamental laser pulse. The third-order polarization is proportional to the third power of the incident electric field: \( P^{(3)} \sim \chi^{(3)} E^3(t) \). The intensity of the unfocused beam is not high enough for this third-order nonlinear conversion. We increase the intensity of the nonlinear interaction by focusing the beam using a telescope. We use a prism to build a crude spectrometer that spatially separates the two beams.

3.1.1 **Set-up of the telescope and the crude spectrometer:**

a) Discuss how to set up a telescope that focuses the beam and then makes it collimated again so that it can be propagated through the prism and onto the scattering cube. How is the beam diameter changed?

b) Set up the telescope. Align one lens at the time using as low power as possible.

c) Align the prism and let the beam propagate onto a white paper on a beam block. Why can we see the third harmonic?

d) Explain why we do not see the 2\textsuperscript{nd} harmonic.

*Hint:* In atomic gases the polarization \( P \) is always parallel to the electric field \( E \) due to the inversion symmetry of the potential: \( V(\vec{r}) = V(-\vec{r}) \).

**Good luck and Enjoy 😊 !!!**