OPTI 511L
Fall 2018

Experiment 3: Second Harmonic Generation (SHG) (1 week lab)

In this experiment we produce 0.53 μm (green) light by frequency doubling of a 1.06 μm (infrared) diode-pumped YAG laser, using a KTP crystal as the nonlinear medium. We will:

A. Demonstrate frequency doubling of a YAG laser (1064 nm -> 532 nm).

B. Observe how the optical power at 532 nm strongly depends on *phase-matching*.

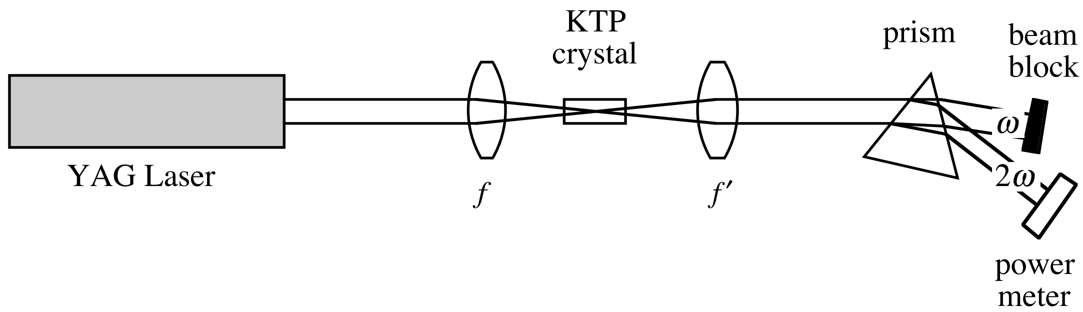
C. Optimize and measure the efficiency of second harmonic generation.

SAFETY NOTE

For this experiment we use an Amoco Model ALC 1064-150P laser, which produces around 150 mW of CW single mode power at 1.064 μm. The human eye is not sensitive to this wavelength, but the radiation is transmitted through the eye and focused near the retina, where it **can cause irreversible damage**. The danger threshold for CW viewing is around 1 mW/cm^2 entering the pupil, so you need to be careful. Protective goggles will be available, and should be used at all times.

Second Harmonic Generation (SHG)

Set up the following (*this may need to be modified further for reliable measurements*!):



1. Start with a lens of focal length f~ 10 or 5 cm. To get a feel for the approximate beam diameter at the focus, assume an approximately 5 mm beam diameter from the 1.064 micron YAG laser, *calculate the beam waist and confocal parameter at the focus (ie confocal parameter = 2 x Rayleigh Range). How does this compare to the length of the 5 mm KTP crystal? What will be the beam waist and confocal parameter for the second harmonic beam? (Keep in mind that the actual beam waist coming from the laser is likely strongly diverging).*

2. The polarization of the YAG laser is linear. Adjust the rotation angle of the KTP crystal such that the polarization of the 1.064 μm beam forms an angle $\theta = 45°$ with its square edges, and so the beam is focused at the center of the crystal. (If the angle of the linear polarization is not known, adjust the KTP to optimize the second harmonic light) Fine adjust the tip and tilt angles of the KTP to maximize the green (532 nm) light. Make sure you also adjust the longitudinal crystal position so that the beam waist coincides with the crystal position. A translation stage will be useful to optimize the alignment.

3. Observe the power of the 532 nm light as a function of the phase matching angle. Try to <u>accurately</u> measure the maximum green light you can generate. *This can be quite challenging because the intense beam at 1.064 µm can easily corrupt your measurement, and the generated light will be very weak*. You may need more than a single prism to separate the beams sufficiently. A color filter may also be used to confirm that the power you measure is truly the green light (532 nm). Think carefully how to do this and make sure to estimate any significant losses from the prisms and/or filters. Also be careful of scattered light (e.g. from the prism), which may be detected on the power meter. Also, test background levels by turning off the room lights. The photodiode can easily detect the room lights if not blocked. You may also need to continually check and correct the alignment of the beams through the crystal and onto the power meter & beam block as you change the phase matching angle.

4. Try a few different focussing lenses (e.g. 5 cm and 10 cm) in order to optimize the SHG conversion efficiency. **Which choice of lens gives you the maximum amount of SHG?** Note that the choice of focal length f involves a tradeoff: a short focal length increases the intensity at the beam waist needed for efficient SHG, but at the same time decreases the Rayleigh range for the beam. If f becomes too short the beam diffracts rapidly, and the intensity does not remain high along the entire length of the crystal.

5. Although the generated SHG light power (532 nm) is low, try to **measure and graph the detected SHG power vs. incident power** (ie you'll need at least 3 points!) How would you expect this graph to look if you were able to take enough points with a good signal? (e.g. a linear or nonlinear relationship?)

6. What is the highest conversion efficiency, η , that you can obtain experimentally? When you determine η , you must account for losses - especially reflection loss at the prism or any uncoated surfaces.

7. Near the angle of optimum second harmonic generation, record the dependence of second harmonic power versus the rotation angle of the crystal (see discussion at end of this document).

Background: Some elements of nonlinear optics

Nonlinear optical susceptibility:

The propagation of a field E through a polarizable medium with polarization density P is governed by the wave equation,

$$\nabla^2 E - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}E = \mu_0 \frac{\partial^2}{\partial t^2}\mathsf{P} \ . \qquad (1)$$

In linear optics we have

$$\mathsf{P} = \varepsilon_0 \chi E ,$$

where the linear susceptibility χ is a (constant) material parameter. When we solve the problem of wave propagation we find simultaneous, self-consistent solutions to these two equations. Note that the linearity of the medium response implies a superposition principle – electromagnetic waves passing through a linear medium do not interact.

In nonlinear optics we make the generalization

$$\mathsf{P} = \varepsilon_0 \chi(E) E = \varepsilon_0 (\chi^{(1)} E + \chi^{(2)} E \cdot E + \chi^{(3)} E \cdot E \cdot E + ...) , \qquad (2)$$

i. e. we allow the medium to respond to the driving field in a nonlinear fashion. We see immediately that the polarization density will contain components $\mathsf{P}^{(2)} \propto E^2$, $\mathsf{P}^{(3)} \propto E^3$ etc. For a monochromatic driving field of frequency ω, these components will radiate at 0ω, ω, 2ω, 3ω etc. The nonlinear susceptibilities are small, $\chi^{(1)} >> \chi^{(2)} >> \chi^{(3)} >> \ldots$, and the conversion into higher harmonics is correspondingly inefficient - except for extremely intense fields.

Note that the nonlinearity brings with it a breakdown of the superposition principle. If the driving field contains two frequencies ω_1 and ω_2 then in addition to higher harmonics we also get sum- and difference frequency generation, $\omega_1 + \omega_2$ and $\omega_1 - \omega_2$. Electromagnetic waves passing through a nonlinear medium interact!

Second harmonic generation:

We now examine the wave equation (1) with a nonlinear polarization (2) up to and including the 2nd order susceptibility. The driving and second harmonic fields have the form

$$E_\omega = \tfrac{1}{2}\mathsf{E}_\omega(z)e^{-i(\omega t - k_\omega t)} + c.c.,$$
$$E_{2\omega} = \tfrac{1}{2}\mathsf{E}_{2\omega}(z)e^{-i(2\omega t - k_{2\omega} t)} + c.c.. \tag{3}$$

To leading orders these waves induce a polarization density

$$\begin{aligned}\mathsf{P} = \mathsf{P}_0^{(NL)}(z) &+ \tfrac{1}{2}\mathsf{P}_\omega^{(L)}(z)e^{-i(\omega t - k_\omega t)} + c.c. \\ &+ \tfrac{1}{2}\mathsf{P}_{2\omega}^{(NL)}(z)e^{-i(2\omega t - 2k_\omega t)} + c.c. \\ &+ \tfrac{1}{2}\mathsf{P}_{2\omega}^{(L)}(z)e^{-i(2\omega t - k_{2\omega} t)} + c.c.\end{aligned} \tag{4}$$

($\chi^{(2)} E \cdot E$ contains a DC term which gives rise to a DC polarization component $\mathsf{P}_0^{(NL)}(z)$, which we will ignore). The linear terms in the polarization can be accounted for in the usual way: at frequencies well away from any absorption resonances they simply add to the (real) index of refraction. We therefore have

$$k_\omega = n_\omega \frac{\omega}{c}, \qquad k_{2\omega} = n_{2\omega}\frac{2\omega}{c}.$$

In general $n_{2\omega} \neq n_\omega$ and therefore $k_{2\omega} \neq 2k_\omega$. Light radiated by the nonlinear polarization at points z and $z + \Delta z$ differs in phase by $e^{-i2k_\omega \Delta z}$, while the 2ω wave propagating from z to $z + \Delta z$ picks up a phase $e^{-ik_{2\omega}\Delta z}$. Clearly then, there is the possibility of a *phase mismatch* between the propagating and generated light, which prevents the two from interfering constructively. This subtle point turns out to be an all-important consideration in determining the efficiency of any nonlinear conversion process.

The wave equation for the second harmonic component is

$$\frac{\partial^2}{\partial z^2} E_{2\omega} - \frac{n_{2\omega}^2}{c^2} \frac{\partial^2}{\partial t^2} E_{2\omega} = \mu_0 \frac{\partial^2}{\partial t^2} \mathsf{P}_{2\omega}^{(NL)} \tag{5}$$

Plugging in the field and nonlinear polarization at 2ω from eqs. (3) and (4), using the slowly varying envelope approximation for $\mathsf{E}_{2\omega}(z)$ and $\mathsf{P}_{2\omega}^{(NL)}(z)$ we obtain

$$\frac{d\mathsf{E}_{2\omega}(z)}{dz} = i\omega \frac{\mu_0 c}{n_{2\omega}} \mathsf{P}_{2\omega}^{(NL)}(z) e^{i(2k_\omega - k_{2\omega})z} = i\omega \frac{\mu_0 c}{n_{2\omega}} \bar{d} \mathsf{E}_\omega(z)^2 e^{i\Delta kz}, \tag{6}$$

where

$$P_{2\omega}^{(NL)}(z) \equiv \bar{d} \mathsf{E}_\omega(z)^2, \quad \Delta k \equiv 2k_\omega - k_{2\omega}.$$

The quantity $\bar{d}$ is a material parameter that characterizes the effective nonlinearity of the medium. For **KTP we have** $\bar{d} \approx 17 \times 10^{-9}$ esu $\approx 6 \times 10^{-23}\,\mathrm{C/V^2}$.

When the power in the second harmonic wave remains much smaller than the power in the fundamental (no depletion approximation) we can set $\mathsf{E}_\omega(z)^2 = \mathsf{E}_\omega(0)^2$ and integrate eq. (6) to find

$$\mathsf{E}_{2\omega}(z) = i\omega \frac{\mu_0 c}{n_{2\omega}} \bar{d} \mathsf{E}_\omega(z)^2 z \frac{\sin(\Delta kz/2)}{\Delta kz/2} e^{i\Delta kz/2}.$$

Using $I_{2\omega}(z) = \frac{1}{2} c\varepsilon_0 |\mathsf{E}_{2\omega}(z)|^2$, and assuming an input intensity I_ω and a doubling crystal of length L, we finally obtain the intensity of the second harmonic wave at the output facet,

$$I_{2\omega}(L) = \kappa I_\omega^2 L^2 \left[\frac{\sin\left(\frac{1}{2}\Delta kL\right)}{\frac{1}{2}\Delta kL} \right]^2. \tag{7}$$

This allows us to estimate the power conversion efficiency,

$$\eta \equiv \frac{P_{2\omega}}{P_\omega} = 2 \left[\frac{\mu_0}{\varepsilon_0} \right]^{3/2} \frac{\omega^2 d^2 L^2}{n^3} \left[\frac{P_\omega}{\pi w^2} \right] \frac{\sin^2(\Delta kL/2)}{(\Delta kL/2)^2}, \tag{8}$$

where P_ω, and $P_{2\omega}$ is the optical power in the fundamental and second harmonic beam, respectively. For simplicity we have approximated $I_\omega \approx P_\omega / 2w^2$, where w is the beam radius of the fundamental. Equations (7) and (8) are our key result. Clearly, good conversion efficiency requires $\Delta k \sim 0$ and large I_ω.

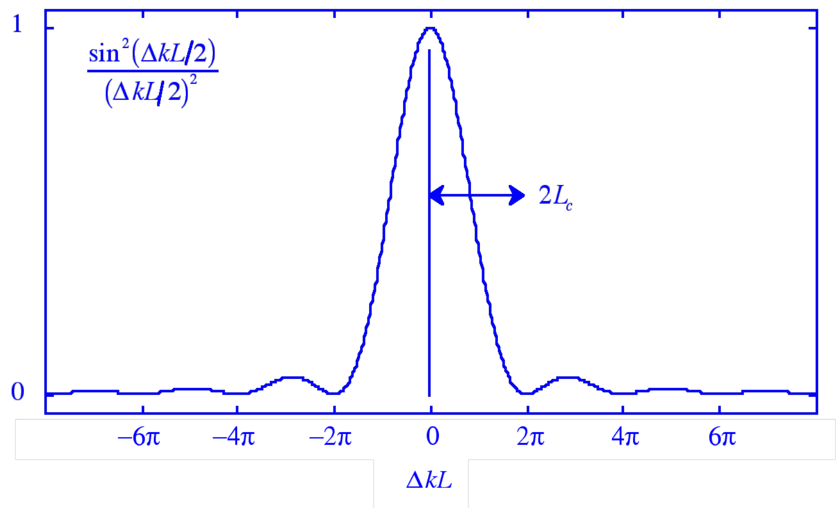


Fig. 1: Second harmonic generation efficiency as function of phase mismatch ΔkL

The *Coherence Length* L_c is defined by

$$|\Delta k| L_c \equiv \pi \;\Rightarrow\; L_c = |\pi/\Delta k|,$$

and sets the length scale over which $E_{2\omega}$ and $\mathsf{P}_{2\omega}$ stays phase matched. Reexamining eq. 7 we see that

$$I_{2\omega}(L) \propto \sin^2\left(\frac{\pi}{2}\frac{L}{L_c}\right),$$

i. e. given a certain minimum Δk the optimum crystal length is L_c. Conversely, for a given crystal length we are doing an adequate job of phase matching as soon as $L_c >> L$.

Phase matching strategies.

There are several ways in which we can independently adjust n_ω and $n_{2\omega}$ to achieve perfect phase matching, $\Delta k = 0$. The most common method takes advantage of the fact that doubling crystals are *birefringent*.

In a *uniaxial* crystal the index of refraction for a wave polarized along one crystal axis is different from the index along the remaining two axes, $n_x \neq n_y = n_z$. Consider then a field propagating along the z' axis, forming an angle φ with the z axis.

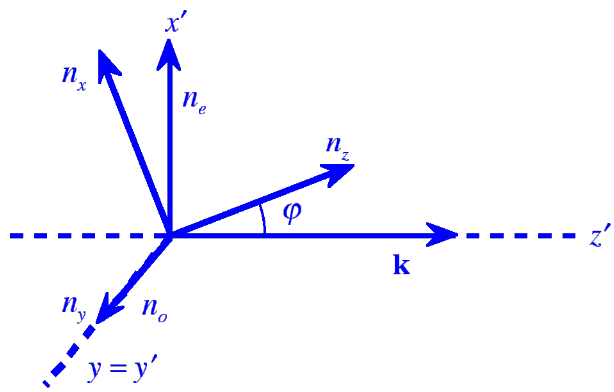


Fig. 2. Indices of refraction in a uniaxial crystal.

In this situation we get extraordinary and ordinary indices of refraction for linear polarizations along x' and y,

$$n_o = n_y\ , \qquad n_e = \frac{n_x^2 + n_z^2}{n_x^2 \cos^2 \varphi + n_z^2 \sin^2 \varphi} \qquad [9]$$

In *angle tuned phase matching* we adjust the angle φ (by rotating the crystal). There are two ways of doing this:

Type I phase matching: $\quad n_{2\omega}^{o} \cdot 2\omega = n_{\omega}^{e} \cdot \omega + n_{\omega}^{e} \cdot \omega \Rightarrow n_{2\omega}^{o} = n_{\omega}^{e}$

Type II phase matching: $\quad n_{2\omega}^{o} \cdot 2\omega = n_{\omega}^{o} \cdot \omega + n_{\omega}^{e} \cdot \omega \Rightarrow n_{2\omega}^{o} = \frac{1}{2}\left(n_{\omega}^{o} + n_{\omega}^{e}\right)$

In Type I phase matching the fundamental is polarized along x' and the second harmonic is polarized along y; two extraordinary photons at ω combine to generate one ordinary photon at 2ω.

In Type II phase matching the fundamental is polarized at $45°$ to the x', y axes and the second harmonic is polarized along y; an ordinary and an extraordinary photon at ω combine to generate an extraordinary photon at 2ω.

Phase matching in our experiment is complicated by the fact that KTP is a *biaxial* crystal, i. e. $n_x \neq n_y \neq n_z$. We will not discuss biaxial phase matching here, but simply note that our crystal has already been cut to achieve Type II phase matching when the laser beam is normal to the entrance facet, and linearly polarized along the diagonal.

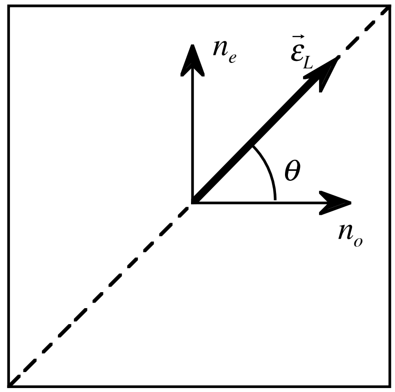


Fig. 3. Phase matching occurs for our precut KTP crystal when the laser polarization $\vec{\varepsilon}_L$ forms an angle $\theta = 45°$ with the ordinary/extraordinary axes.

Since only Type II phase matching can occur in our crystal, the second harmonic field $E_{2\omega} \propto E_\omega^o E_\omega^e$, where $E_\omega^o \propto E_\omega \cos(\theta)$ and $E_\omega^e \propto E_\omega \sin(\theta)$. This immediately tells us that

$$I_{2\omega} \propto \left[I_\omega \cos(\theta)\sin(\theta)\right]^2 \propto I_\omega^2 \sin^2(2\theta).$$

As our crystal is rotated around the axis of the laser beam, the second harmonic intensity will therefore change as a function of θ, with a period of $90°$.

References:

"Lasers", P. W. Milonni and J. H. Eberly (Wiley 1988).
"Quantum Electronics", 2nd Ed., A. Yariv (Wiley 1975).
"Nonlinear Optics", R. W. Boyd (Academic Press 1992).