



# Research-Group Internship

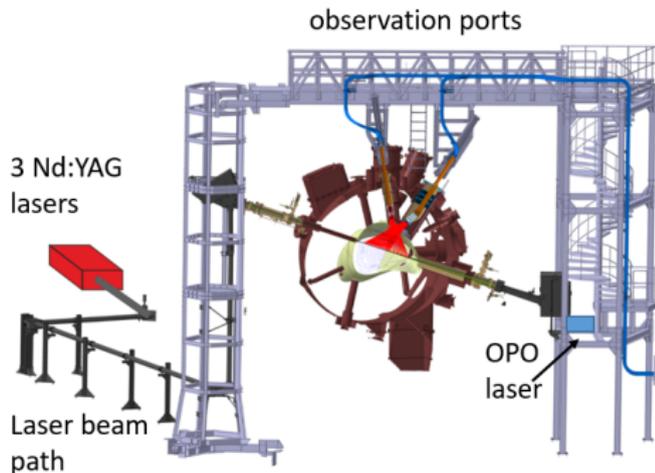
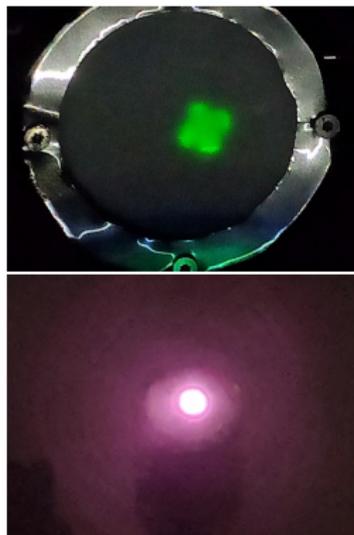
## Understanding the emitted green light of the Thomson Nd:YAG lasers

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4th of August, 2023

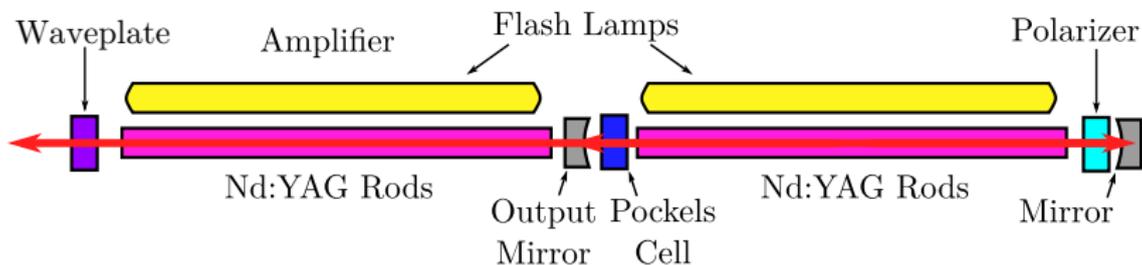
# Internship Reasons and Goals



Thomson Scattering beam path

Images of diffuser under 1064 nm laser light, with (b) and without (t) 1064 nm filter.

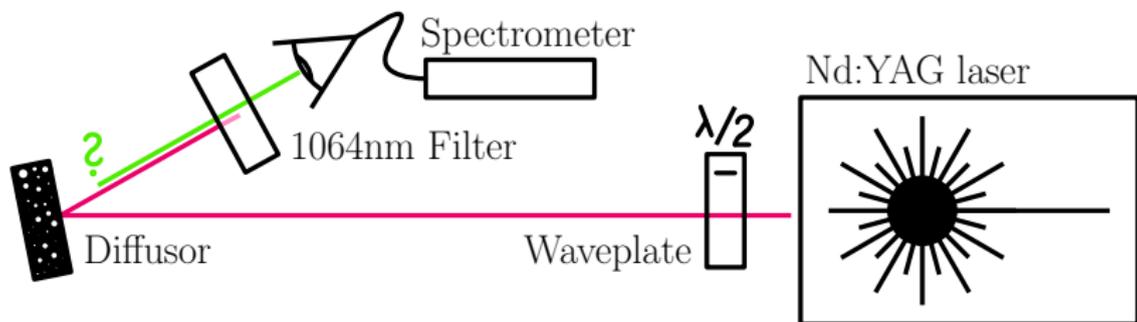
# The Nd:YAG laser



Nd:YAG schematic

- four level laser, 1064 nm
- polarized output light
- experiment lasers rotate polarization via waveplate
- pulsed with 10 ns and 1.8 J

# Spectrum Measurements



Experimental setup for the frequency distribution measurement

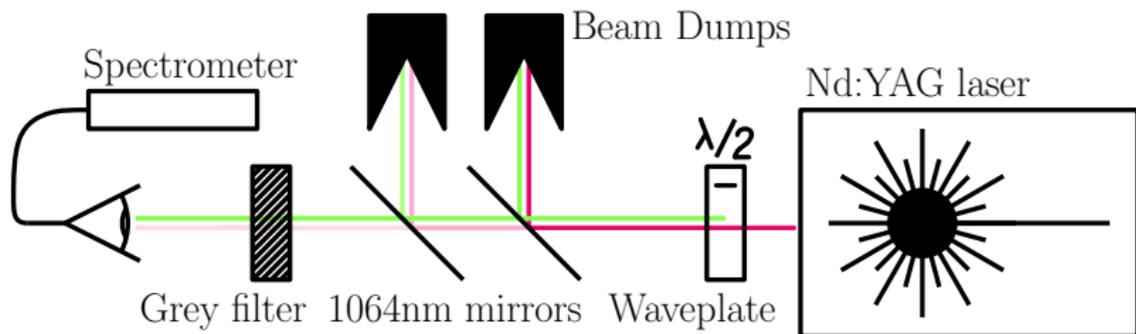
- goal: measure spectrum of the emitted green light to gather information about its cause
- spectrum measurement of scattered light



- tested 1064 nm experiment lasers, varying power of green light
- switch to laboratory 1064 nm laser during the experiment  
campaign no green light
- sanity check of visible light generation from original  
lasers after campaign easily visible green light

results not reproducible

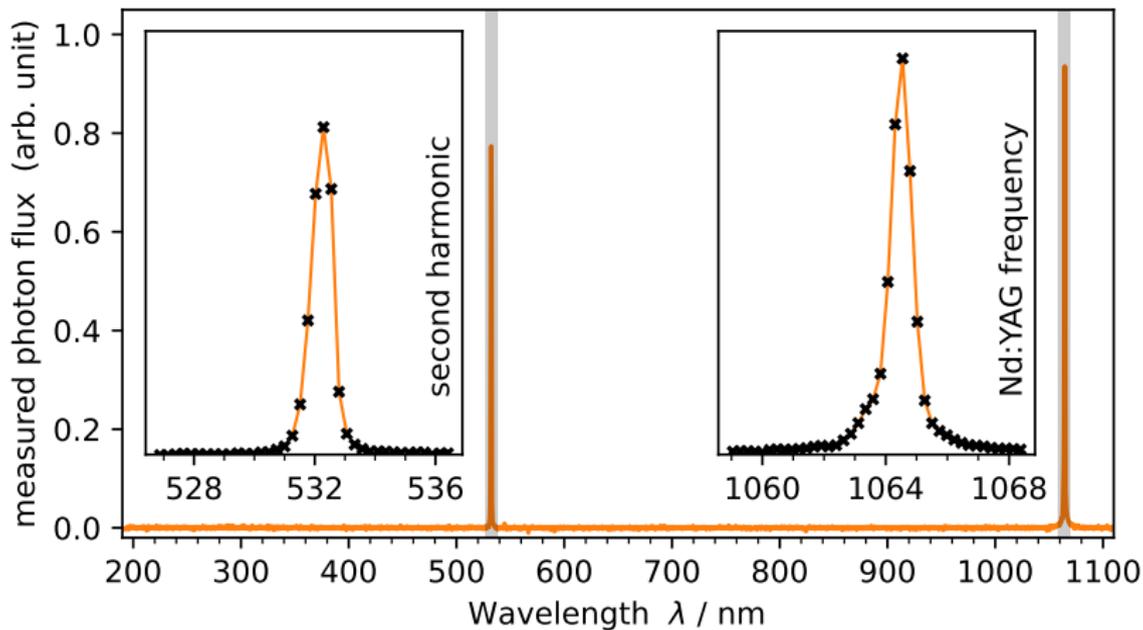
only viable difference: quartz  $\lambda/2$ -waveplate



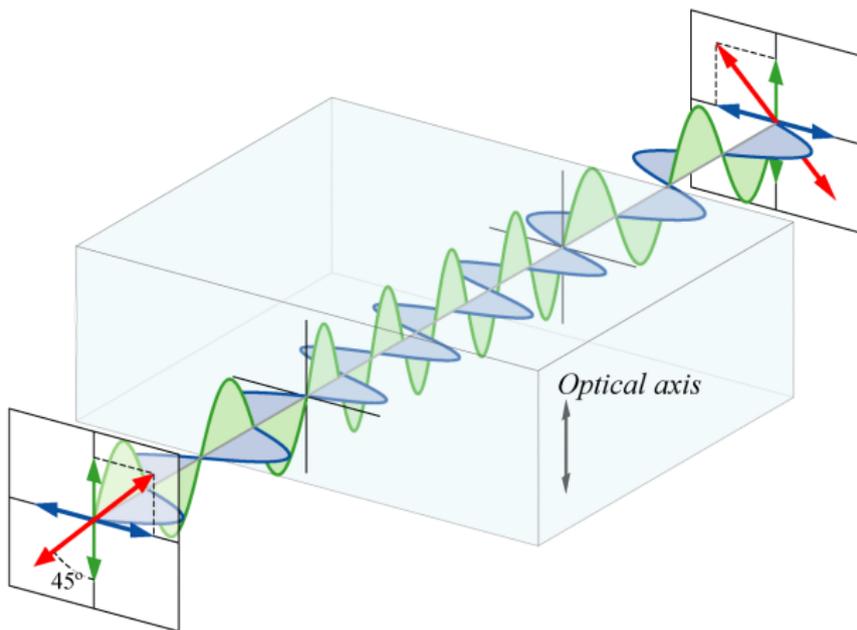
New experimental setup for the frequency distribution measurement

- new hypothesis: generation of green light in waveplate
- spectrum measurement of unscattered light possible
- 1064 nm light filtered after waveplate to reduce energy influx

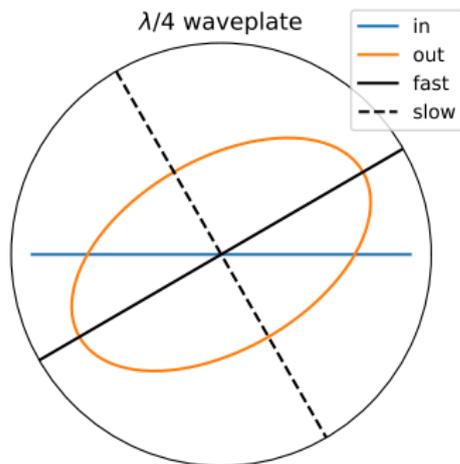
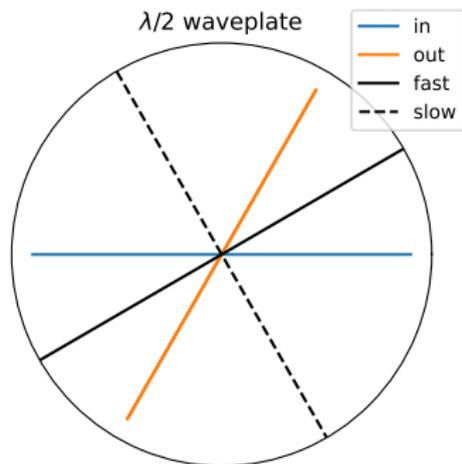
# Spectrum Measurements



Frequency distribution measurement of the visible green light

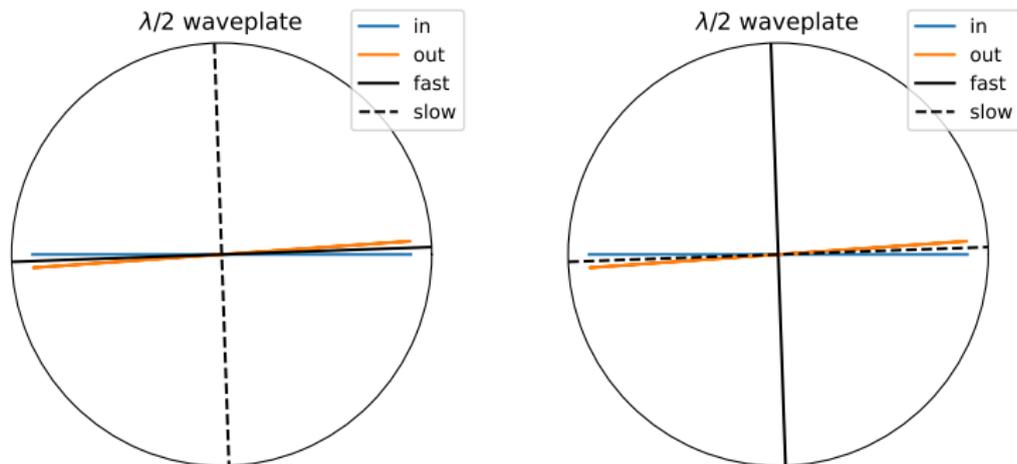


$\lambda/2$  – waveplate (positive uniaxial medium)



phase changes due to  $30^\circ$  rotated waveplates ( $\Delta\varphi_{\text{left}} = \pi$ ,  $\Delta\varphi_{\text{right}} = \pi/2$ )

Given  $\Delta\varphi(L) = \frac{2\pi}{\lambda}L = \pi$ , and a angle  $\alpha$  between the input polarization and the fast axis, the polarization is rotated by  $2\alpha$ .



phase changes due to  $\approx 0^\circ$  and  $\approx 90^\circ$  rotated waveplates ( $\Delta\varphi = \pi$ )

Given  $\Delta\varphi(L) = \frac{2\pi}{2} = \pi$ , and a angle  $\alpha$  between the input polarization and the fast axis, the polarization is rotated by  $2\alpha$ .



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FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 Å. The arrow at 3472 Å indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 Å is very large due to halation.

First published SHG spectrum

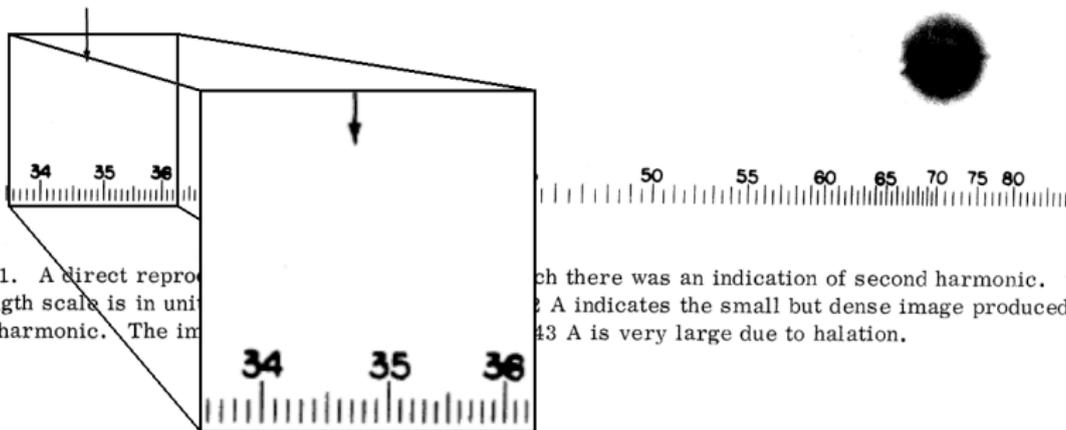
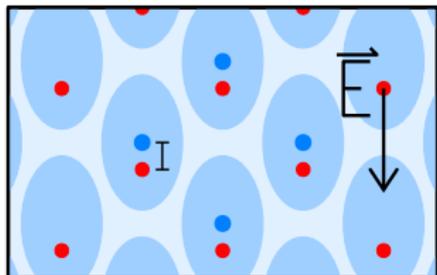
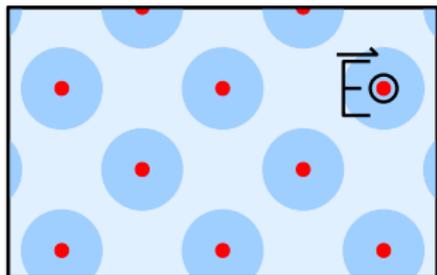


FIG. 1. A direct reproduction of the first published SHG spectrum. The wavelength scale is in units of the fundamental wavelength. The small spot at 34.3 A is the second harmonic. The large spot at 34.3 A is very large due to halation.

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First published SHG spectrum

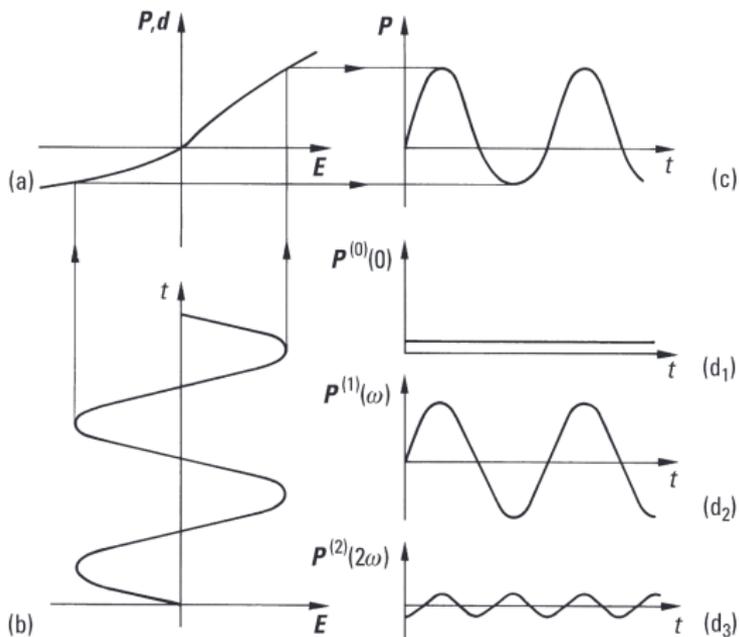


Polarization in crystals

- $\vec{E}$  field polarizes crystal
- $\vec{P} = P_0 + \varepsilon_0 \chi^{(1)} \vec{E} + \mathcal{O}(\vec{E}^2)$

However

- $\vec{P}$  dependent on unit cell choice  
→  $\Delta \vec{P}$  independent
- $\chi$  frequency dependent
- $\vec{P} \propto \vec{E}$  only holds for small  $\vec{E}$  in isotropic materials



Effect of 1D  $\vec{P}(\vec{E})$  nonlinearities



$$E(t) = E_0 \sin(\omega t)$$

non-pyroelectric (Quartz):

$$\frac{P(E)}{\epsilon_0} = \frac{P_0}{\epsilon_0} + \chi^{(1)} E + \chi^{(2)} E^2 + \mathcal{O}(E^3) \quad P(0) = 0 \implies P_0 = 0$$

$$\begin{aligned} \implies \frac{P(t)}{\epsilon_0} &= \frac{P_0}{\epsilon_0} + \chi^{(1)} E_0 \sin(\omega t) + \chi^{(2)} E_0^2 \sin^2(\omega t) \\ &= \left( \frac{P_0}{\epsilon_0} + \frac{\chi^{(2)} E_0^2}{2} \right) + \chi^{(1)} E_0 \sin(\omega t) - \frac{\chi^{(2)} E_0^2}{2} \cos(2\omega t) \end{aligned}$$

$\implies$  SHG for 2nd order terms in  $P(E)$



$$E(t) = E_0 \sin(\omega t)$$

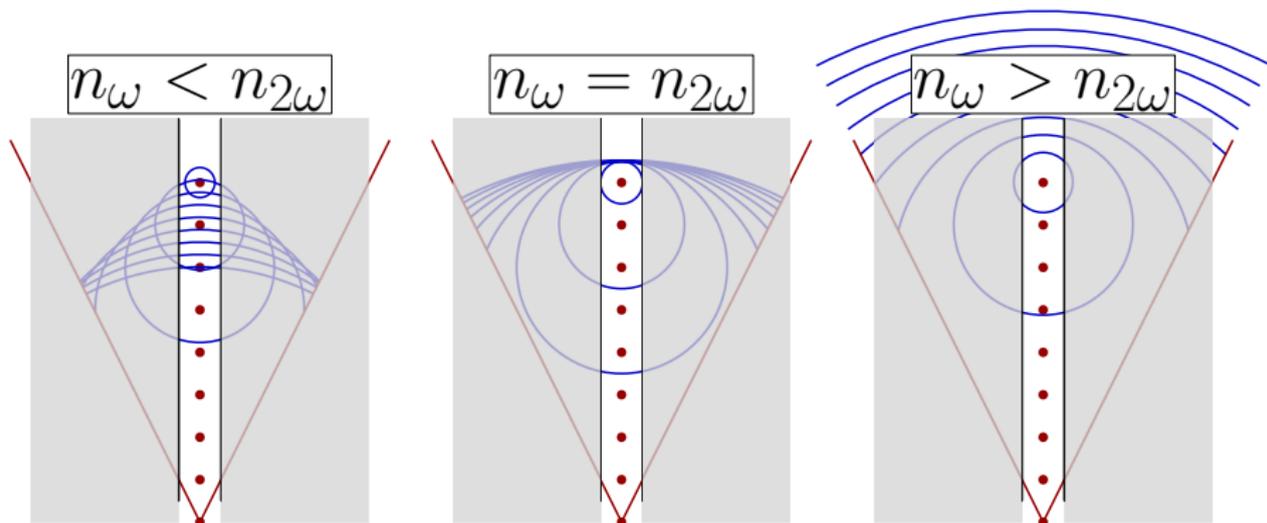
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$$\sin^n(\omega t) = 2^{1-n} \begin{cases} \frac{1}{2} \binom{n}{\frac{n}{2}} + \sum_{k=1}^{\frac{n}{2}} \binom{n}{\frac{n}{2} - k} (-1)^k \cos(\underline{2k}\omega t) & n \text{ even} \\ \sum_{k=0}^{\frac{n-1}{2}} \binom{n}{\frac{n-1}{2} - k} (-1)^k \sin(\underline{(2k+1)}\omega t) & n \text{ odd} \end{cases}$$

# Intro to SHG

## Macroscopic Scale



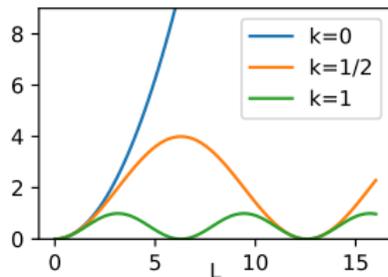
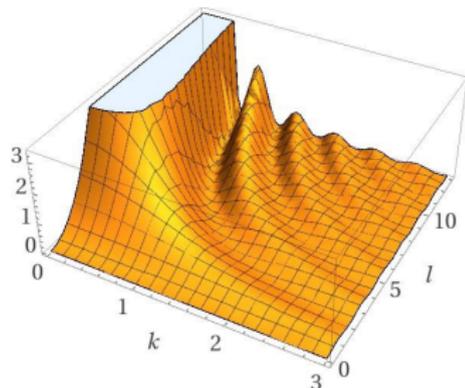
SHG interference at and near phase-matched conditions

$$\Delta k = k_{2\omega} - 2k_{\omega} = \frac{2\omega}{c} (n_{2\omega} - n_{\omega}) = \frac{2\omega}{c} \Delta n$$

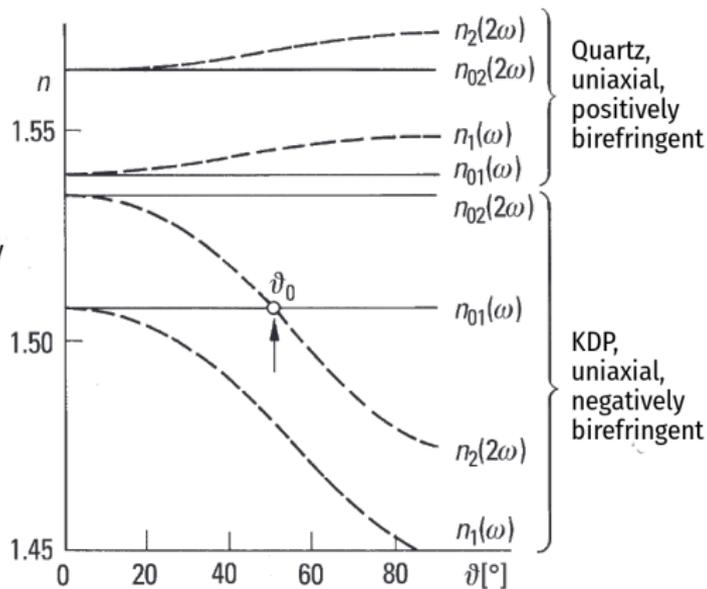
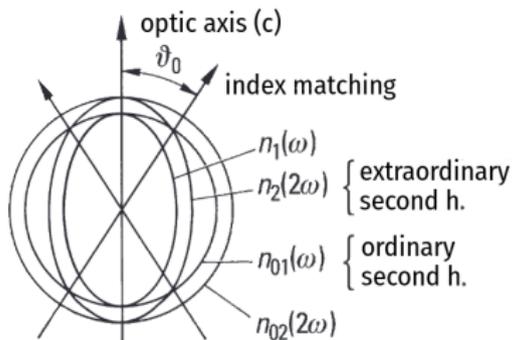


$$I_{2\omega}(l) \propto \left[ \frac{\sin(\Delta k l/2)}{\Delta k} \right]^2 d_{\text{eff}}^2 \omega^2 I_{\omega}^2$$
$$\propto \left[ \frac{\sin(\Delta n l \omega / c)}{\Delta n} \right]^2$$

- minimize  $\Delta n$
- for  $\Delta n > 0$ :
  - $\hat{I} \propto \Delta n^{-2}$
  - $\lambda_I = \frac{\pi c}{\Delta n \omega}$



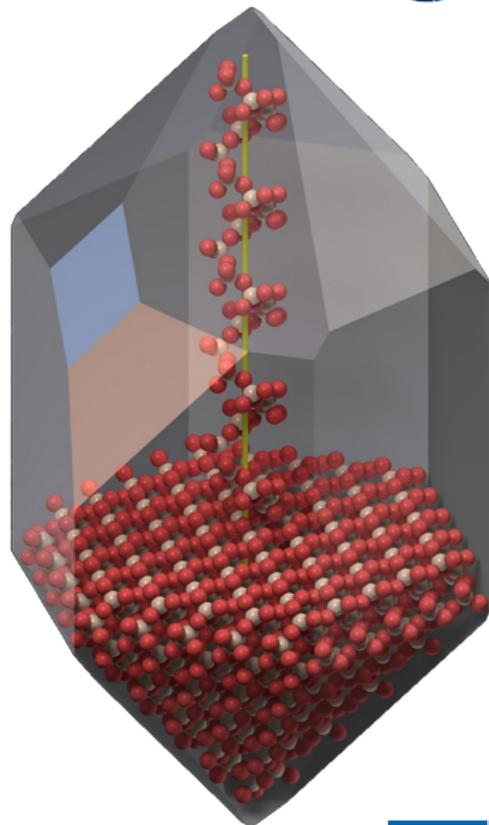
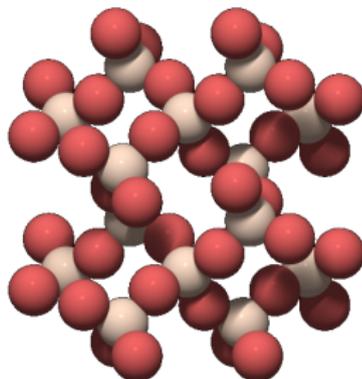
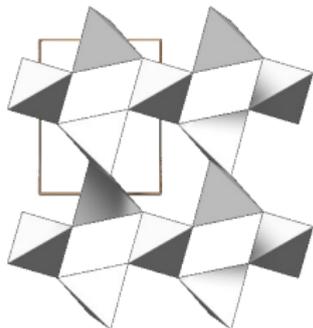
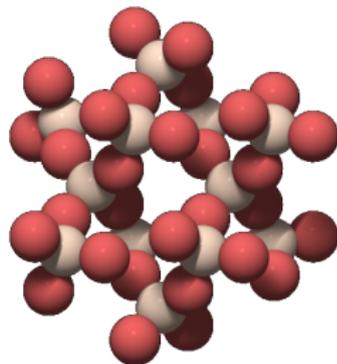
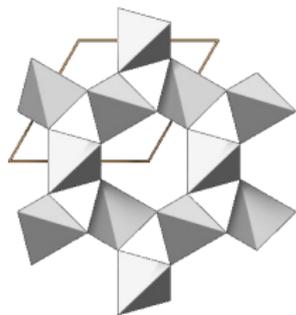
$$\left[ \frac{\sin(kl/2)}{k} \right]^2$$



Index matching concept for KDP and quartz. Quartz not possible:

$$[n_o(\omega), n_{eo}(\omega)] = [1.534, 1.543] < [1.547, 1.556] = [n_o(2\omega), n_{eo}(2\omega)]$$

# Quartz and its Properties



# Quartz and its Properties

## Electric Susceptibility Tensors

$P_0 = 0$  as quartz is non-pyroelectric

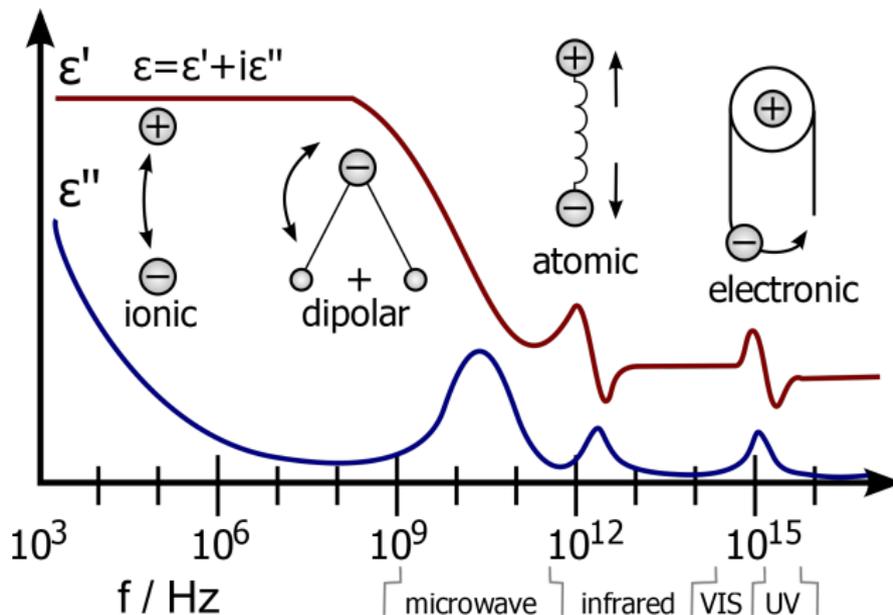


# Quartz and its Properties

## Electric Susceptibility Tensors



$$P_0 = 0$$



Quartz Structure

# Quartz and its Properties

## Electric Susceptibility Tensors



$$P_0 = 0, \quad \chi^{(1)} \approx 1.5$$

$$\varepsilon_r^0 \approx \begin{pmatrix} 4.64 & & \\ & 4.64 & \\ & & 4.85 \end{pmatrix} \quad \varepsilon_r^\infty \approx \begin{pmatrix} 2.51 & & \\ & 2.51 & \\ & & 2.55 \end{pmatrix}$$

$$\begin{aligned} \implies \chi^{(1)} = \varepsilon_1 - 1 &\approx 3.7 \text{ in the DC region} \\ &\approx 1.5 \text{ in the NIR and VIS region} \end{aligned}$$

# Quartz and its Properties

## Electric Susceptibility Tensors



$$P_0 = 0, \quad \chi^{(1)} \approx 1.5$$

$$\vec{P}_\mu^{(2)}(t) = \varepsilon_0 \iint_{-\infty}^{\infty} \chi_{\mu\alpha\beta}^{(2) \text{ sum}}(\omega_1, \omega_2) \vec{E}_\alpha(\omega_1) \vec{E}_\beta(\omega_2) e^{-i(\omega_1 + \omega_2)t} d\omega_1 d\omega_2$$

assuming frequency independence of  $\chi$  for SHG,  
and monofrequent input EM-waves ( $\omega_1 = \omega_2 = \omega$ )

$$\vec{P}_\mu^{(2)}(2\omega) = \varepsilon_0 d_{\mu\alpha\beta} \vec{E}_\alpha(\omega) \vec{E}_\beta(\omega)$$

$$d, \chi^{(2)} \in \mathbb{C}^{3 \times 3 \times 3} \quad d_{\mu\alpha\beta} := \frac{1}{2} \chi_{\mu\alpha\beta}^{(2)}$$

# Quartz and its Properties

## Electric Susceptibility Tensors

$$P_0 = 0, \quad \chi^{(1)} \approx 1.5, \quad \varepsilon_0 \vec{E}_\alpha \vec{E}_\beta d_{\mu\alpha\beta} = \vec{P}_\mu^{(2)}$$

$d \in \mathbb{C}^{27}$  in general



# Quartz and its Properties

## Electric Susceptibility Tensors

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$d \in \mathbb{C}^{27}$  in general

$\mathbb{C}^{18}$  including symmetry in  $\alpha$  and  $\beta$

$$(d_{\mu\alpha\beta} = d_{\mu\beta\alpha})$$

$$\begin{array}{cccccc} \alpha\beta = & xx & yy & zz & yz & zx & xy \\ \mu = x & \left( \begin{array}{cccccc} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{array} \right) & = & d_{\mu[\alpha\beta]} \\ & y & & & & & \\ & z & & & & & \end{array}$$



# Quartz and its Properties

## Electric Susceptibility Tensors



$$P_0 = 0, \quad \chi^{(1)} \approx 1.5, \quad \varepsilon_0 \vec{E}_\alpha \vec{E}_\beta d_{\mu\alpha\beta} = \vec{P}_\mu^{(2)}$$

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$$(d_{\mu\alpha\beta} = d_{\alpha\mu\beta})$$

$$\begin{array}{l} \alpha\beta = xx \quad yy \quad zz \quad yz \quad zx \quad xy \\ \mu = x \begin{pmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{31} & d_{21} \\ y \begin{pmatrix} d_{21} & d_{22} & d_{23} & d_{32} & d_{14} & d_{12} \\ z \begin{pmatrix} d_{31} & d_{32} & d_{33} & d_{23} & d_{13} & d_{14} \end{pmatrix} \end{pmatrix} \end{pmatrix} = d_{\mu[\alpha\beta]} \end{array}$$

# Quartz and its Properties

## Electric Susceptibility Tensors



$$P_0 = 0, \quad \chi^{(1)} \approx 1.5, \quad \varepsilon_0 \vec{E}_\alpha \vec{E}_\beta d_{\mu\alpha\beta} = \vec{P}_\mu^{(2)}$$

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$\mathbb{R}^2$  Neumann's point group principle  $(d_{\mu\alpha\beta} = (Td)_{\mu\alpha\beta})$

$$\mu = \begin{matrix} \alpha\beta = & xx & yy & zz & yz & zx & xy \\ x & \left( d_{11} & \overline{d_{11}} & & d_{14} & & \right) \\ y & & & & & \overline{d_{14}} & \overline{d_{11}} \\ z & & & & & & \end{matrix} = d_{\mu[\alpha\beta]}^{(32) \text{ sym.}}$$

# Quartz and its Properties



## Electric Susceptibility Tensors

$$P_0 = 0, \quad \chi^{(1)} \approx 1.5, \quad \varepsilon_0 \vec{E}_\alpha \vec{E}_\beta d_{\mu\alpha\beta} = \vec{P}_\mu^{(2)}$$

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$\mathbb{R}^1$  Neumann's principle & Kleinmann's rule

$$\mu = \begin{matrix} \alpha\beta = & xx & yy & zz & yz & zx & xy \\ x & \left( d_{11} & \overline{d_{11}} & & & & \right) \\ y & & & & \overline{d_{11}} & & \\ z & & & & & & \end{matrix} = d_{\mu[\alpha\beta]}^{(32)}, \text{ no loss}$$

# Quartz and its Properties



## Electric Susceptibility Tensors

$$P_0 = 0, \quad \chi^{(1)} \approx 1.5, \quad \varepsilon_0 \vec{E}_\alpha \vec{E}_\beta d_{\mu\alpha\beta} = \vec{P}_\mu^{(2)}, \quad d_{11} \approx 0.3 \text{ pm/V}$$

$d \in \mathbb{C}^{27}$  in general

$\mathbb{C}^{18}$  including symmetry in  $\alpha$  and  $\beta$   $(d_{\mu\alpha\beta} = d_{\mu\beta\alpha})$

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$$\mu = \begin{matrix} \alpha\beta = & xx & yy & zz & yz & zx & xy \\ x & \left( d_{11} & \overline{d_{11}} & & & & \right) \\ y & & & & \overline{d_{11}} & & \\ z & & & & & & \end{matrix} = d_{\mu[\alpha\beta]}^{(32)}, \text{ no loss}$$

# Quartz and its Properties



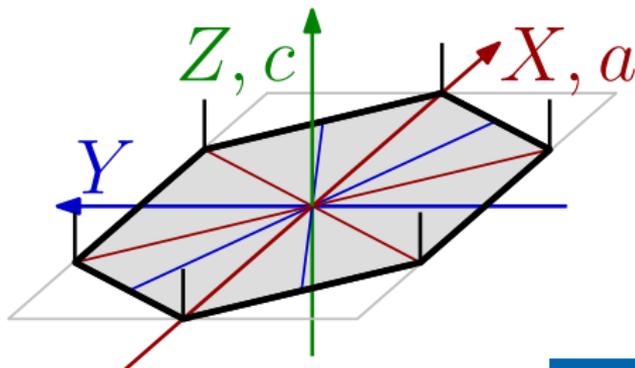
## SHG Direction Dependence

$$d_{11} = -d_{12} = -d_{26} \approx 0.3 \text{ pm/V} \quad \varepsilon_0 \vec{E}_\alpha \vec{E}_\beta d_{\mu\alpha\beta} = \vec{P}_\mu^{(2)}$$

$$\Rightarrow \frac{\vec{P}(2\omega)}{d_{11}\varepsilon_0} = \frac{1}{d_{11}\varepsilon_0} \begin{pmatrix} P_x \\ P_y \\ P_z \end{pmatrix} = \begin{pmatrix} E_x^2 - E_y^2 \\ -2E_x E_y \\ 0 \end{pmatrix} = \begin{pmatrix} \cos 2\theta \\ -\sin 2\theta \\ 0 \end{pmatrix} E^2 = E^2 R_{2\theta} \hat{e}_x$$

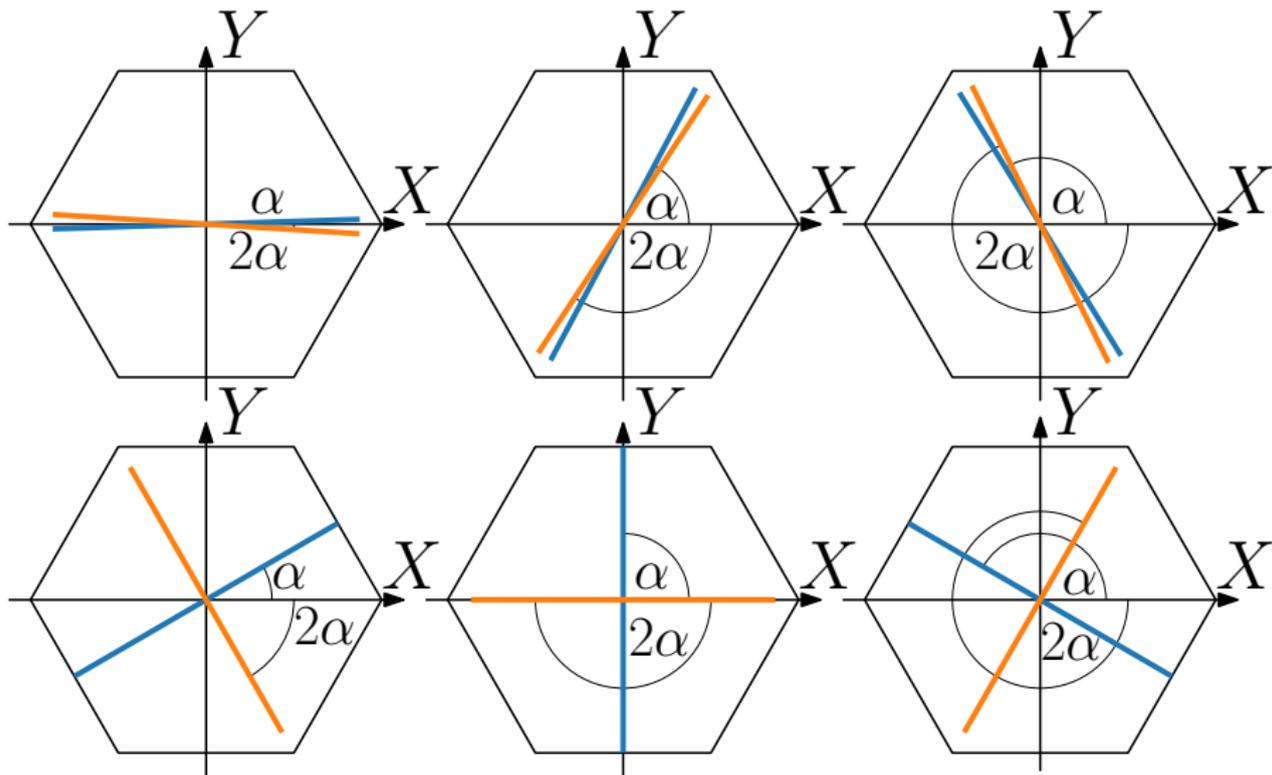
with  $\vec{E}$  rotating in the XY-plane with magnitude E and angle  $\theta$  to  $\hat{e}_x$   
Thus, for  $E_z = 0$ :

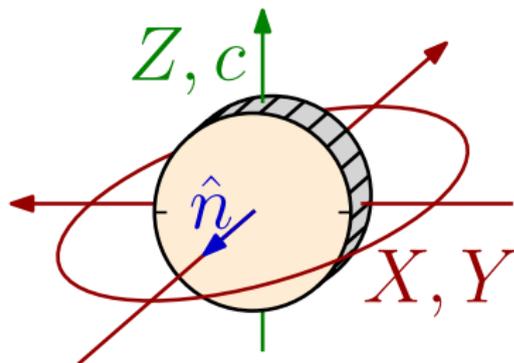
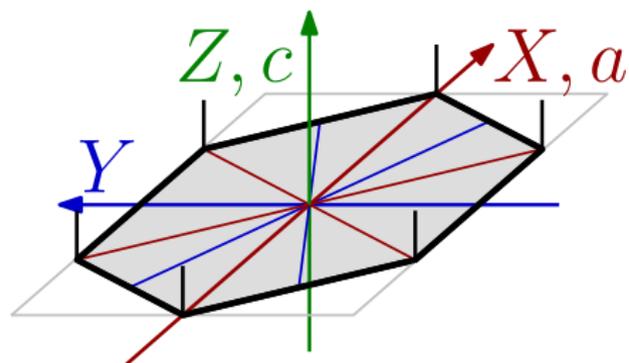
$$\vec{E}^2 \propto \|\vec{P}\| \not\propto f(\theta)$$



# Quartz and its Properties

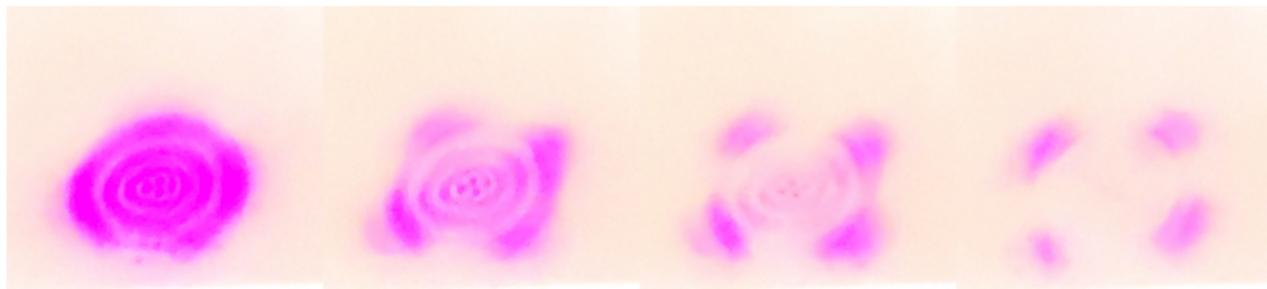
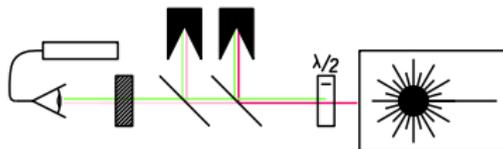
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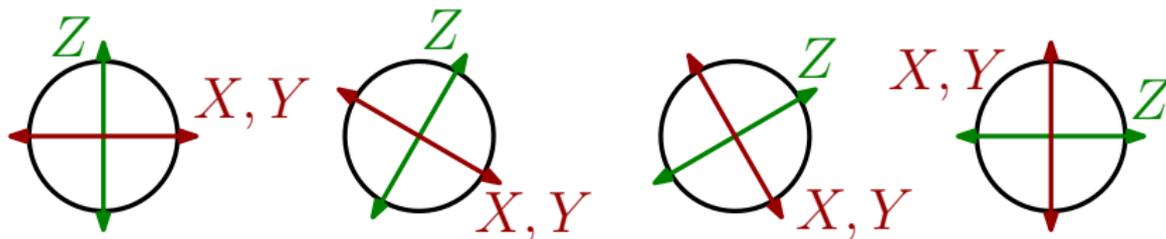


- laser light along  $\hat{n}$  polarized in  $XY$ -plane and angle  $\theta = \angle \hat{e}_x \vec{E}$   
 $\Rightarrow P_{\perp \hat{n}} = \|\vec{P} \times \hat{n}\| \propto \|\cos 2\theta \cos \theta - \sin 2\theta \sin \theta\| = \|\cos 3\theta\|$ 
  - for  $\vec{E} \parallel \hat{e}_x$ , SHG along  $\hat{n}$  is maximized
  - for  $\vec{E} \parallel \hat{e}_y$ , SHG along  $\hat{n}$  is minimized
- no phase matching possible, but for our laser,  $I(2\omega)/I(\omega) \sim 10^{-7}$   
non phase-matched intensity is enough

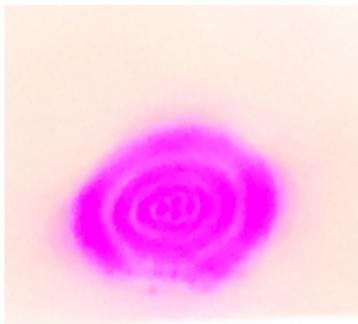
# Effects in our Waveplate



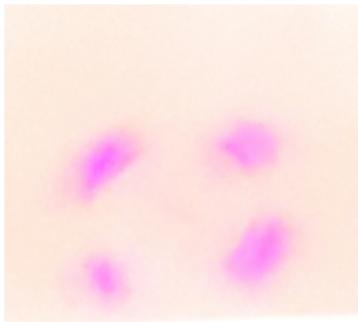
visible SHG at rotation angles  $\Delta\alpha \approx 0^\circ, 30^\circ, 60^\circ$  and  $90^\circ$ . (inverted)



# Effects in our Waveplate

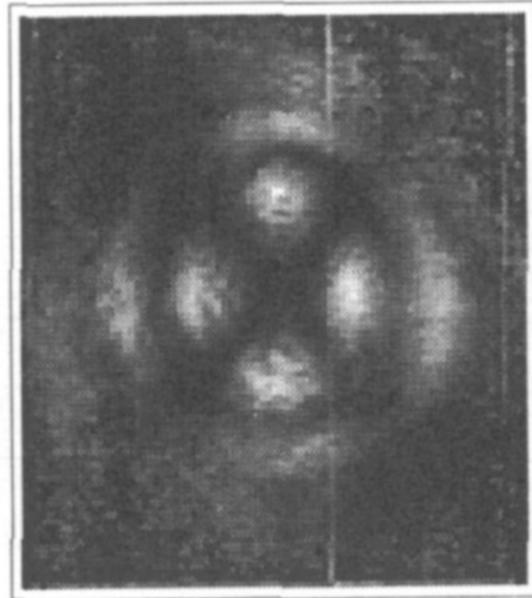
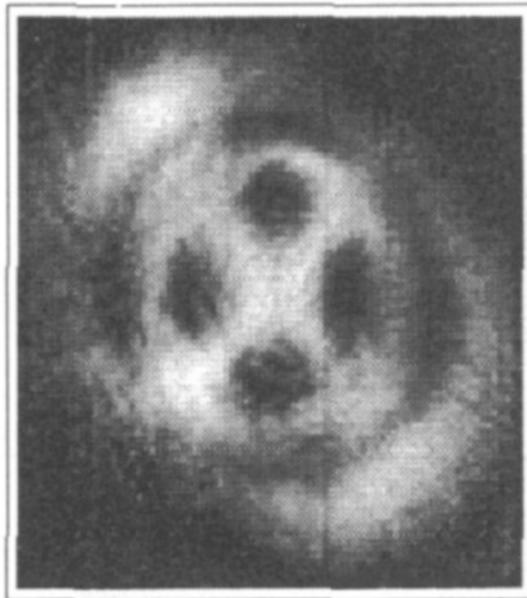


- $\vec{E}_{\text{out}}(\omega) \parallel \vec{E}_{\text{in}}$ , unchanged, horizontally pol.
  - $\vec{E}_{\text{out}}(2\omega) \parallel \vec{E}_{\text{in}}$ , mostly horizontally pol.
- Nd:YAG light frequency doubled



- $\vec{E}_{\text{out}}(\omega) \parallel \vec{E}_{\text{in}}$ , unchanged, horizontally pol.
  - $\vec{E}_{\text{out}}(2\omega) \perp \vec{E}_{\text{in}}$ , mostly vertically pol.
- depolarized Nd:YAG light frequency doubled

# Nd:YAG Depolarization



Captured Nd:YAG light passing through a polarization filter polarized parallel (left) and perpendicular (right) to the intended laser polarization



The goal of the internship was achieved:

- quartz waveplate is responsible for green light instead of diffusor
- green light is second harmonic (532 nm) of Nd:YAG wavelength
- the theory on SHG and waveplates can explain the observed light and its intensity distribution can be explained by

⇒ Possible combined SHG+waveplates for use in laser adjustment

- improvement in adjustment visibility, with only diffusive elements needed as detector cards
- non-hazardous adjustment together with filter for fundamental wavelength

# References I



[A.C. Akhavan, quartzpage.de/gen\\_struct.html](http://quartzpage.de/gen_struct.html).  
Quartz structure, 2017.  
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# Research-Group Internship

## Understanding the emitted green light of the Thomson Nd:YAG lasers

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4th of August, 2023