Exercises Nonlinear Optics 2018

The following sections of the book "Nonlinear optics by R. W. Boyd, Third Edition, will be treated in the current course:

Chapter 1, paragraphs: 1.1 – 1.6
Chapter 2, paragraphs: 2.1 – 2.5
Chapter 4, paragraphs: 4.1 – 4.3; part 4.4, 4.5, 4.6
Chapter 11, paragraphs: 11.1-11.3
Chapter 12, paragraphs: 12.1-12.4

The precise homework assignment for the following week will be communicated during the course.

List of constants:

Bohr radius (a_o):	5.29177 x 10^{-11} m
Elementary charge, e:	1.6 x 10^{-19} C
The dielectric permittivity of free space:	8.85 x 10^{-12} C^2 J^{-1} m^{-1}
Mass of an electron, m:	9.1 10^{-31} kg
Dielectric constant of vacuum:	1
Boltzmann constant, k:	1.38 x 10^{-23} m^2 kg s^{-2} K^{-1}
Avogadro number, N_A:	6.022 x 10^{23} mol^{-1}
Density of liquid hydrogen (33 K):	70.85 kg/m^3
Heat capacity per unit volume of water (ρ_0C at 25 °C):	4.2 J/cm^3 K
Thermal conductivity of water (κ, 25 °C):	0.56 W /m K
Dielectric constant of water, ε:	80 (298 K)
Dipole moment of water:	1.85 D
1 D:	3.336 10^{-30} Cm
Radius of a water molecule:	0.14 nm
Absorption coefficient of water, α (~632 nm):	0.6 m^{-1}
Excitation wavelength of electrons in water:	180 nm

Refractive index change with temperature for water:

$-\frac{dn}{dT} = a(1 - e^{-\frac{(T-T_0)}{T_k}})$; valid in the range [– 3, 80] °C;
with a =2.62·10^{-5} K^{-1}; T_0 = 2.0 °C T_k= 48.5 °C

Useful expressions:

Taylor Series expansion $f(x) = \sum_{n=0}^{\infty} \frac{f(0)^{n'}}{n!} x^n$

Trigonometric identities:

$\sin x = \frac{e^{ix} - e^{-ix}}{2i}$; $\cos x = \frac{e^{ix} + e^{-ix}}{2}$

$\sin 2x = 2 \sin x \cos x$
$\cos 2x = \cos^2 x - \sin^2 x$

$\cos(x \pm y) = \cos(x) \cos(y) \mp \sin(x) \sin(y)$
$\sin(x \pm y) = \sin(x) \cos(y) \pm \cos(x) \sin(y)$

Definition of the delta function:

$\delta(x - a) = \int_{-\infty}^{\infty} e^{-i(x-a)t} dt$ and consequently $f(x = a) = \int_{-\infty}^{\infty} \delta(x - a) f(x) dx$

Laser intensity: $I = 2n\epsilon_0 c E^2$

Fresnel factors:
E_0 incoming wave
E'_0 transmitted wave
E''_0 reflected wave
n, μ: incoming medium
n', μ': outgoing medium
i: angle with surface normal incoming medium
polarization: $\perp$ to plane of incidence

$$\frac{E'_0}{E_0} = \frac{2n \cos i}{n \cos i + \frac{\mu}{\mu'} \sqrt{n'^2 - n^2 sin^2 i}}$$

$$\frac{E''_0}{E_0} = \frac{n \cos i - \frac{\mu}{\mu'} \sqrt{n'^2 - n^2 sin^2 i}}{n \cos i + \frac{\mu}{\mu'} \sqrt{n'^2 - n^2 sin^2 i}}$$

Nonlinear Optical Processes

Nonlinear optics is a sub-branch of optics that describes the optical response of materials when the interaction of the optical field results in an optical field whose amplitude depends nonlinearly on the amplitude of the incoming field. Such processes occur when the field strength of the incoming light is of the same order of magnitude as the electrical field that exists between the electrons and the nuclei of a material. Nonlinear optical phenomena have been predicted since the 1930's and were observed experimentally when laser began to be used as light sources in the early 1960's. With the technological advancements over the last decades resulting in lasers that can produce intense fields with ultrashort pulse lengths and tunable wavelengths, nonlinear optical phenomena have become commonplace in optical devices and materials.

Nonlinear optical processes

1.
a. Starting from a beam with the following frequency (ω) components $E(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + c.c$, compute explicitly the second order components of the nonlinear polarization $P^{(2)}(t) = \epsilon_0 \chi^{(2)} E(t)^2$ and write down what each frequency component represents. Make a list of nonlinear optical phenomena that you know of for each frequency component.
b. Which processes do you know for $E(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + E_3 e^{-i\omega_3 t} + c.c$ and $P^{(3)}(t) = \epsilon_0 \chi^{(3)} E(t)^3$. There is no need to write this out explicitly.

The atomic polarizability

2.
In order to estimate the electronic polarizability of an atom and derive some of its linear properties, we consider the simplest possible model of an atom, the Bohr hydrogen (H) atom. In this model the H atom is represented by a charged nucleus with mass M and charge +e. The electron orbits the nucleus in a spherical shell. The situation is depicted in Fig 1a.

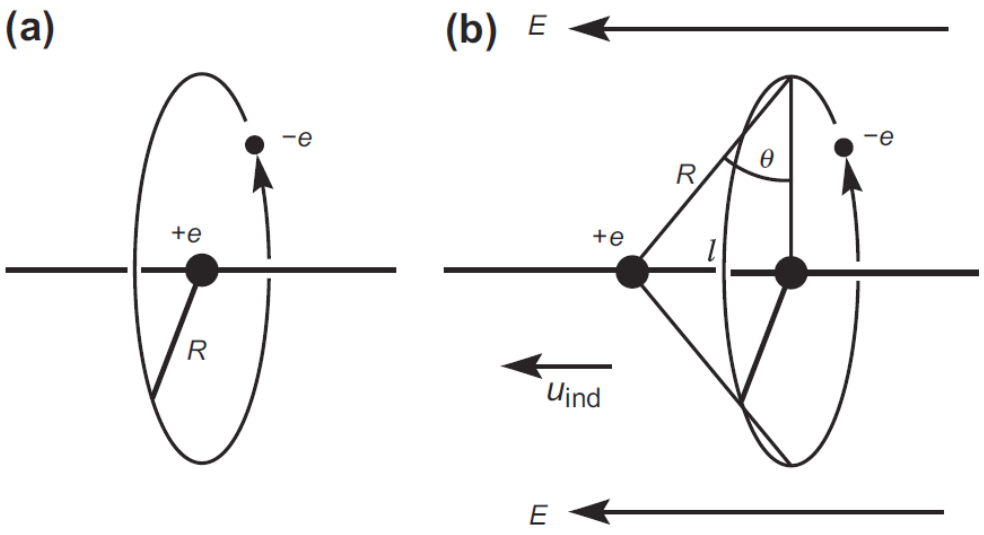

Figure 1: Model of the Bohr atom, without and with an electric field (E). Note that we do not use the symbol ***u*** but ***p*** for the induced dipole moment.

a. In this model, which forces make sure that the electron keeps its steady orbit?

When an external electric field (*E*) is present that can be considered as very weak compared to the intra-atomic field (from a neighboring charge for example) the atom will be distorted in shape until the forces are again in balance (Fig 1b).
b. Which part of the atom is actually distorted?

In the thus distorted atom a dipole moment is induced that can be written as: $p_{ind} = \alpha_0 E$
with α_0 the electronic polarizability of the atom.

c. Starting from the expression for the force balance, show that $\alpha_0 = 4\pi\varepsilon_0 R^3$. For the H atom R takes the value of the Bohr radius.
d. To understand the influence of many such atoms, we consider the case of liquid hydrogen. Calculate the dielectric constant of liquid hydrogen.
e. Based on the expressions for the induced dipole moment determine the units for the macroscopic polarization, and determine if that is a useful set of units.

Electric field inside an atom and at an interface
3.
a. Using the picture in Fig. 1 determine the electric field strength in a typical atom.
b. Calculate the intensity that is needed to reach the magnitude of an atomic field, and determine if such a condition can be reached with a 1 W 100 fs laser source with a variable repetition rate from 1 kHz to 1 MHz focused down to 100 μm^2.
c. At a typical interface between media there is often a layer of charge separation. The interface possesses a surface potential that decays over a short distance. Typically 150 mV decays are achieved within 1 nm. What is the electrostatic field strength? Do you expect that a nonlinear interaction can occur with it?

The properties of the nonlinear optical susceptibility
4.
We have seen that by definition there are a large number of nonlinear susceptibility elements.
a. In theory, how many elements of $\chi^{(3)}$ are permitted if we want to satisfy the condition $\omega_4 = \omega_1 + \omega_2 + \omega_3$?
b. How many different tensor elements remain if we require that all fields present in the medium are real (and thus measurable) properties?
c. How many different tensor elements are left if we are dealing with a process that is lossless?
d. How many different tensor elements are left if we also can omit effects of dispersion (i.e. Kleinman's symmetry)?

5.
For the second-order susceptibility many studies assume Kleinman's symmetry in the analysis of the outcome of an experiment or to describe crystal properties. As a consequence the number of nonzero and independent tensor elements is reduced and it is possible to introduce a contracted notation. In this notation the following definition is made:

$$\chi_{ijk}^{(2)} = 2d_{il}$$

with i representing the first index and l representing the pair j.k as follows:

jk:	11,	22,	33,	23=32,	13=31,	12=21
l:	1,	2,	3,	4,	5,	6

a. Show that there are 18 possible elements using this notation.
b. Show that for the case when Kleinman's symmetry holds there are only 10 independent nonzero elements.

Spatial Symmetry

6. First-order susceptibility
For monoclinic crystals (with point group C_2) we have seen that the non-zero tensor elements for the first-order nonlinear susceptibility are: xx,yy,zz,xy,yx (with z being the symmetry axis)
a. Show that for orthorhombic crystals (with a C_{2v} point group; meaning there is a C_2 axis, and two orthogonal reflection planes that contain the C_2 axis) two further elements will vanish. Which are those?
b. Show how further restrictions come into play for tetragonal crystals that have an additional C_4 axis along the z direction.
c. Cubic crystals with point group O have 3 C_4 axes in addition to being orthorombic. What are the consequences?
d. What are the implications for an isotropic medium?

7. Second-order susceptibility
The nonlinear optical crystal KDP belongs to the space group D_{2d} or $\bar{4}2m$. This space group has among others the following symmetry operations:

$$D(S_4) = \begin{pmatrix} 0 & -1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & -1 \end{pmatrix}; D(C_2) = \begin{pmatrix} -1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 1 \end{pmatrix}; D(\sigma_d) = \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}$$

a. Determine the non-zero elements of the second-order nonlinear susceptibility of KDP.

Cubic crystals with point group O have 3 C_4 axes.
b. Which second-order susceptibility elements are non-zero for such cubic crystals?
c. Using this result, which second-order susceptibility elements are non-zero for isotropic materials?

When we perform second-order experiments between isotropic materials the nonlinear optical processes report primarily on the interfacial structure. To describe this structure it makes sense to define the second-order surface polarizability, here given for SHG:
$P_{s,i}^{(2)}(2\omega) = \epsilon_0 \chi_{s,ijk}^{(2)}(2\omega, \omega, \omega) E_j(\omega) E_k(\omega)$,
with s the subscript to indicate the surface.
d. What should be the definition of second-order surface susceptibility $\chi_{s,ijk}^{(2)}(2\omega, \omega, \omega)$ and what would be the units for $\chi_{s,ijk}^{(2)}(2\omega, \omega, \omega)$ and $P_{s,i}^{(2)}(2\omega)$?
e. If the interface is charged, we saw earlier that it may be possible that the electrostatic field in the electric double layer can participate in the nonlinear optical process. What susceptibility tensor element will mediate this interaction in a second harmonic experiment? Assume that the material is lossless. What form will the nonlinear polarizability have? Assume that the electrostatic field has a penetration depth of ~ 20 nm into the material. We need to know the response of the entire surface region.

Tensor properties resulting from the anharmonic oscillator model

8.
Starting from the expression obtained for a collection of isotropic, anharmonic oscillators:

$$\chi^{(3)}_{ijkl}(\omega_q;\omega_m,\omega_n,\omega_p) = \frac{Nbe^4(\delta_{ij}\delta_{kl}+\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk})}{3\epsilon_0 m^3 D(\omega_q)D(\omega_m)D(\omega_n)D(\omega_q)}$$

a. Show that the nonlinear susceptibility possesses the following tensor properties:
$\chi_{1122} = \chi_{1212} = \chi_{1221} = \chi_{1133} = \chi_{1313} = \chi_{1331} = \chi_{2233} = \chi_{2323} = \chi_{2332} = \chi_{2211} = \chi_{2121} = \chi_{2112} = \chi_{3311} = \chi_{3131} = \chi_{3113} = \chi_{3322} = \chi_{3232} = \chi_{3223} = 1/3\ \chi_{1111} = 1/3\ \chi_{2222} = 1/3\ \chi_{3333}$
with all other elements vanishing.
b. Give a simple physical argument that explains why the vanishing elements do vanish.
c. Give a simple physical argument that explains why χ_{ijkl} possesses off-diagonal tensor components, even though the medium is isotropic.

Generation of new frequencies in a crystal and Phase matching
9.
We have seen that the wave equation for nonlinear optical processes is of the following form:

$$\nabla^2 \boldsymbol{E}_i(\boldsymbol{r}) + \frac{\omega_i^2}{c^2}\boldsymbol{\epsilon}^{(1)}(\omega_i)\cdot\boldsymbol{E}_i(\boldsymbol{r}) = -\frac{\omega_i^2}{\epsilon_0 c^2}\boldsymbol{P}_i^{NL}(\boldsymbol{r})$$

We seek to find a solution for the case that we have three beams with frequencies ω_3, ω_2 and ω_1 that propagate collinearly along the z direction in a nonlinear crystal which has a single value of $\chi^{(2)}$. ω_3 has no intensity at the entrance to the crystal and is a consequence of the mixing process $\omega_3 = \omega_2 + \omega_1$. We can assume that all fields are plane waves of the form:

$E_i(z,t) = A_i(z)e^{i(k_i z - \omega_i t)}$ + c.c. and the source term is of the form:
$P_i(z,t) = P_i(z)e^{i(k_i z - \omega_i t)}$ + c.c.

a. Find solutions for the field intensities at the exit of the crystal. As an intermediate result you should obtain the following relations for the amplitudes A_1, A_2, A_3:

$$\frac{dA_1}{dz} = \frac{i\chi^{(2)}\omega_1^2}{k_1 c^2}A_3 A_2^* e^{-i\Delta kz}$$
$$\frac{dA_2}{dz} = \frac{i\chi^{(2)}\omega_2^2}{k_2 c^2}A_3 A_1^* e^{-i\Delta kz}$$
$$\frac{dA_3}{dz} = \frac{i\chi^{(2)}\omega_3^2}{k_3 c^2}A_1 A_2\, e^{i\Delta kz}$$

Use for the intensity: $I_i = 2n_i\epsilon_0 c|A_i|^2$. Assume there is no depletion.

For the simple case of phase matching in a nonlinear crystal with just one value of the susceptibility tensor we derived the following expression for the emitted intensity of the third beam (ω_3,I_3):

$$I_3(\omega_3) = \frac{\omega_3^2\left|\chi^{(2)}\right|^2 I_1 I_2 L^2}{2\epsilon_0 c^3 n_3 n_2 n_1} sinc^2\left(\frac{\Delta kL}{2}\right)$$

b. Show that the units are indeed correct for this expression. Compare this result to Eq. 2.2.19 in Boyd (Ed3, page 78). Is anything missing in Eq. 2.2.19?

c. What do we obtain for $\Delta kL \rightarrow 0$. At which values for L will there be no energy conversion into ω_3?

Now we want to do some practical examples. For a negative uniaxial crystal, consider type I collinear phase matching for an SHG process. For this type of crystal we have an index ellipsoid with minimum ($\widetilde{n_e}$) and maximum (n_o) refractive index values. Rotating the crystal over an angle θ, we can tune n:

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

To perform SHG in the crystal we choose three angles θ (0, 45 and 90 degrees) between the optic axis (c) and the **k** vector of the beam.
d. Sketch the crystal with the optic axis, the beam and the best choice for the polarization direction for the incoming and outgoing beam. What value of n does the exiting SH field experience?
e. Using the phase matching condition, derive an expression for the crystal rotation that would provide phase matching.
f. The working range of many crystals is limited. Based on the answer in e, give two reasons for that.

Next, we want to quantify phase matching for the case of second harmonic generation from an interface and in a bulk medium, as is for example the case in SHG imaging or spectroscopy experiments. Use as input parameters: An input beam of 1000 nm wavelength and the refractive index is 1 (air) or 1.33 for (1000 nm) and 1.34 for the second harmonic wave in water.
g. Consider the air / water interface or water as a bulk medium and calculate the distance over which the phase mismatch between the fundamental and second harmonic beams has become π. To obtain the answer sketch the situation for a transmission and a reflection experiment. For simplicity we can assume normal incidence.
h. Why does the SH imaging literature mainly contain images in transmission direction?

The Manley Rowe relations

10.
We have seen in the previous exercise for the derivation of phase matching in a lossless medium that conservation of momentum is a very important requirement. We should also have conservation of energy, which we will calculate here.
a. Calculate for each beam the change in intensity over distance, i.e. $\frac{dI_i}{dz}$. You can use the intermediate result of question 9a.
b. Show that the spatial variation of the total intensity is 0, and describe its meaning.
c. Show that the equality holds:

$$\frac{d}{dz}\left(\frac{I_1}{\omega_1}\right) = \frac{d}{dz}\left(\frac{I_2}{\omega_2}\right) = -\frac{d}{dz}\left(\frac{I_3}{\omega_3}\right)$$

Intensity dependent refractive index

11.
For the third order susceptibility that is responsible for the intensity dependent refractive index we have, for an isotropic material, the following relations:

$\chi_{1122} = \chi_{1133} = \chi_{2233} = \chi_{2211} = \chi_{3311} = \chi_{3322}$;
$\chi_{1212} = \chi_{1313} = \chi_{2323} = \chi_{2121} = \chi_{3131} = \chi_{3232}$;
$\chi_{1221} = \chi_{1331} = \chi_{2332} = \chi_{2112} = \chi_{3113} = \chi_{3223}$;
$\chi_{1111} = \chi_{2222} = \chi_{3333}$;
$\chi_{1111} = \chi_{1122} + \chi_{1221} + \chi_{1212}$;
a. Show that for third harmonic generation these relationships reduce to the one mentioned in question 8.
b. Show that also in the case of Kleinman's symmetry only a single element remains regardless of the number of different frequencies involved.
c. Show that for the frequency combination relevant for the intensity dependent refractive index the induced third-order polarizability will have two components with a different polarization.

12.
The change in the refractive index due to third order processes can originate from a number of different sources, including non-resonant electronic effects and thermal effects. We want to make a comparison for these two effects in liquid water at 25 °C and determine under which conditions either is more important and what parameters are involved.
We have seen that the following expression can be derived from the anharmonic oscillator model.

$$\chi^{(3)}_{ijkl}(\omega_q;\omega_m,\omega_n,\omega_p) = \frac{Nbe^4(\delta_{ij}\delta_{kl}+\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk})}{3\epsilon_0 m^3 D(\omega_q)D(\omega_m)D(\omega_n)D(\omega_q)} \qquad (1)$$

and $D(\omega_i) = \omega_0^2 - \omega_i^2 - 2i\gamma\omega_i$. b is the force constant for the nonlinear interaction. Assume that a good value for b can be found at a distance d when the total restoring force vanishes. d can be approximated by the molecular diameter. In the following we assume that a single beam with a linear polarization interacts with the medium.

a. Show that for this case the non-resonant process results in a single $\chi^{(3)}$ element that has the form:

$$\chi^{(3)} = \frac{Ne^4}{\epsilon_0 m^3 \omega_0^6 d^2} \qquad (2)$$

b. Calculate $\chi^{(3)}$ and n_2 for H_2O.

For a thermal process we can make use of the heat transport equation, from which the laser induced time dependent temperature T_l can be derived:

$$\rho_0 C \frac{dT_l}{dt} - \kappa\nabla^2 T_l = \alpha I \qquad (3)$$

Here, we consider the case of a cylindrical laser beam with a diameter ϕ *that* hits a glass cell with liquid water. The water is heated to a steady state temperature (T_l). Then the beam is blocked and the temperature decays back to its original value in a typical time τ.

c. We see that the equation consists of a source, an uptake part and a transport part. Describe the meaning of each term.

For a very simplistic evaluation we can assume that $\frac{dT_l}{dt} \rightarrow \frac{\Delta T_l}{\Delta t} \sim \frac{T_l}{\tau}$ whereby τ is a typical time constant that characterizes the change in the process. Likewise, we can assume that $\nabla^2 T_l \rightarrow \frac{T_l}{(\phi/2)^2}$
d. Using Eq. 3 show that we have for τ:

$$\tau = \frac{\rho_0 C \phi^2}{4\kappa}$$

e. Calculate the diameter of the illuminating beam that corresponds to τ = 1 μs, 1 ms, 1 s and explain the trend between τ and ϕ.

For a pulsed laser beam with intensity I, diffusion will likely not play a role during the pulse duration τ_p.

f. Determine how single pulsed heating simplifies Eq. 3 and show using the crude simplification we have applied before in part d that for the maximum temperature increase within one pulse we have:

$$T_l^{max} = \frac{\alpha I \tau_p}{\rho_0 C}$$

g. Calculate the maximum temperature increase in liquid water induced by a single 100 fs laser pulse that has a fluence of 10 mJ/cm^2.

h. In part e we saw that the heat dissipation time typically depends on the beam diameter and can vary wildly. Consider for the values τ = 1 μs, 1 ms, 1 s what would be the temperature increase if we had single pulses with a fluence as given in e but originating from different light sources with repetition rates of 1 GHz, 1 MHz and 1 kHz.

Thus, we find that for both a non-resonant electronic process and a heat induced process we have a modification of the refractive index of the form $n = n_0 + \Delta n$. We want to examine for which pulse duration we can neglect thermal effects with respect to electronic effects. We have for thermal effects: $|\Delta n| = \left|\left(\frac{dn}{dT}\right)\right| T_l^{max}(I)$, and $\Delta n = \frac{3\chi^{(3)}}{4\epsilon_0 c n_0^2} I$ for non-resonant electronic effects.

i. Calculate for a laser beam interacting with liquid water the minimum pulse duration for which thermal effects become dominant.

Linear electro-optic effect

13.

Figure 2 shows a picture of a laser system that is regeneratively amplified. The incoming light consists of 800 nm, 88 MHz, 50 fs pulses delivered with a 100 mW power. We seek to convert that into 800 nm, 4 W, 1 kHz pulses with the aid of a 1 kHz, 532 nm, 150 ns, 15 W pump laser. The laser consists of a stretcher that converts the pulses into ~ 100 ps pulses, a regenerative amplifier (RGA) which converts the repetition rate to 1 kHz and increases the pulse energy, a multi-pass amplifier (MPA), which increases the pulse energy and a compressor which converts the pulse back to ~ 70 fs. Both amplification stages use a Ti:Sapphire crystal which convert 532 nm into 800 nm by means of stimulated emission. From the stretcher the pulses hit a polarizer that reflects vertical (V) polarized light and transmits horizontally polarized light (H).The V polarized pulses that travel from the left to the right are rotated in their polarization state to H polarized pulses by a Faraday rotator coil.

a. Describe the operation of the RGA cavity, and the role of each optical element in it. Draw a sketch of the situation and consider the correct timing of the operation of the Pockels Cell (PC).
b. If we connect an oscilloscope to the photodiode (PD) what will we see on the screen if:

1. The 800 nm seed beam is blocked?
2. The cavity runs as it should? Is the timing different from 1?

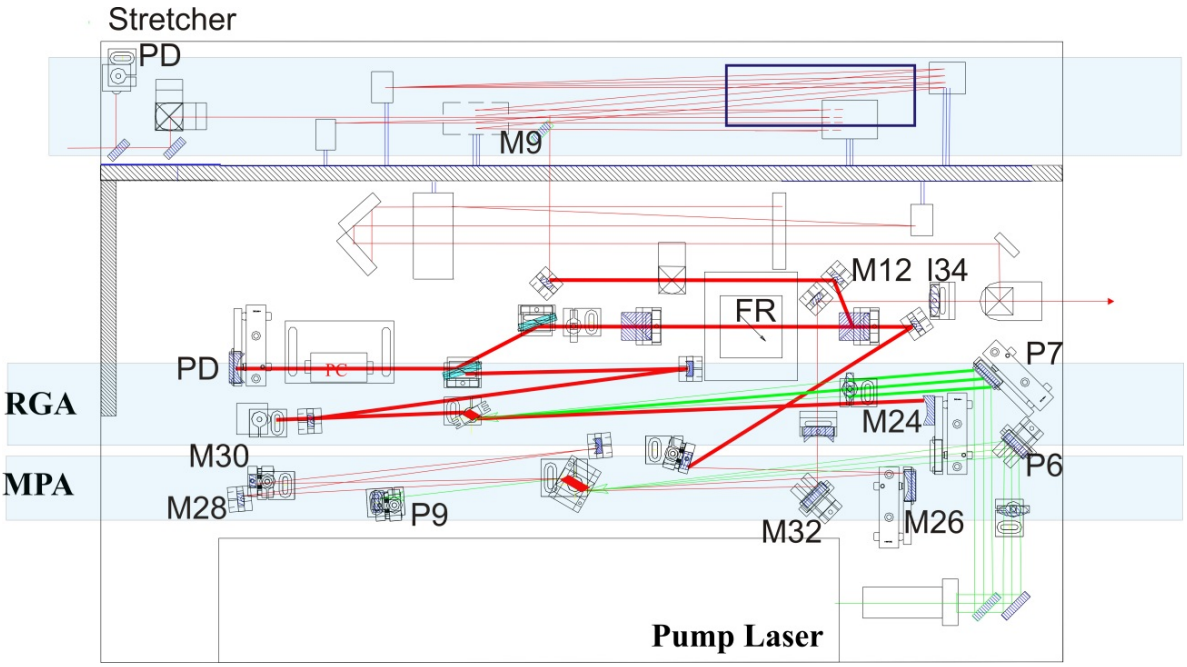

Figure 2: Sketch of an amplified Ti:Sapphire laser. The thicker lines are the important ones relevant for the question. PD: photodiode, PC: Pockels cell, M: seed mirror, P pump mirror, I lens, FR: faraday rotator.

Optical Damage

14.
In a laser system we have a calcite crystal placed in a cavity. A collimated laser pulse with high intensity passes through the crystal.
a. Where in the crystal may we expect to find damage first? Support your answer with a calculation.
b. Which part of a birefringent polarizer is most prone to damage?