FRIEDRICH-SCHILLER-UNIVERSITÄT JENA
PHYSIKALISCH-ASTRONOMISCHE-FAKULTÄT

FRIEDRICH-SCHILLER-
UNIVERSITÄT
JENA

PROF. GERHARD PAULUS

Experimental Nonlinear Optics

All Exercises

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Name: MARTIN BEYER

Contents

1 First Exercise

1.1 Electric Susceptibility for a lossless medium

Show that $\varepsilon_{ij}(\omega)$ is a symmetric tensor for a lossless medium.

Solution: We start by using POYNTINGS theorem of conservation of energy for the electromagnetic field, which can be written in differential form as:

$$-\frac{\partial u}{\partial t} = \vec{\nabla}\cdot\boldsymbol{S} + \boldsymbol{J}\cdot\boldsymbol{E}, \tag{1.1}$$

whee $\vec{\nabla}\cdot\boldsymbol{S}$ is the divergence of the POYNTING vector (energy flow) and $\boldsymbol{J}\cdot\boldsymbol{E}$ is the density of electric power dissipated by the LORENTZ force acting on charge carriers. The term u denotes the energy density which is defined as

$$u = \frac{1}{2}(\boldsymbol{E}\cdot\boldsymbol{D} + \boldsymbol{B}\cdot\boldsymbol{H}). \tag{1.2}$$

For a lossless medium the dissipation term vanishes and we get

$$\vec{\nabla}\cdot\boldsymbol{S} + \frac{\partial u}{\partial t} = 0. \tag{1.3}$$

At first, lets discuss the divergence of the POYNTING vector. For that we use the general relation $\vec{\nabla}\cdot(\boldsymbol{A}\times\boldsymbol{B}) = -\boldsymbol{A}(\vec{\nabla}\times\boldsymbol{B}) + \boldsymbol{B}(\vec{\nabla}\times A)$

$$\begin{aligned}\vec{\nabla}\cdot\boldsymbol{S} &= -\boldsymbol{E}(\vec{\nabla}\times H) + \boldsymbol{H}(\vec{\nabla}\times\boldsymbol{E})\\ &= -\boldsymbol{E}\frac{\partial}{\partial t}\boldsymbol{D} - \boldsymbol{H}\frac{\partial}{\partial t}\boldsymbol{B}\\ &= -E_j\varepsilon_{jk}\frac{\partial}{\partial t}E_k - \mu_0\boldsymbol{H}\frac{\partial}{\partial t}\boldsymbol{H}.\end{aligned} \tag{1.4}$$

Now lets calculate the time derivative of the energy density

$$\frac{\partial u}{\partial t} = \frac{1}{2}\Big(\frac{\partial}{\partial t}E_j\varepsilon_{jk}E_k + E_j\varepsilon_{jk}\frac{\partial}{\partial t}E_k + \underbrace{\mu_0\Big(\frac{\partial}{\partial t}\boldsymbol{H}\Big)\boldsymbol{H} + \mu_0\boldsymbol{H}\frac{\partial}{\partial t}\boldsymbol{H}}_{2\mu_0\boldsymbol{H}\frac{\partial}{\partial t}\boldsymbol{H}}\Big). \tag{1.5}$$

Comparing the last term of (1.5) with (1.4) shows that the last term vanishes in the summation. Therefore the sum can be written as

$$\begin{aligned}E_j\varepsilon_{jk}\frac{\partial}{\partial t}E_k &= \frac{1}{2}\Big(\frac{\partial}{\partial t}E_j\varepsilon_{jk}E_k + E_j\varepsilon_{jk}\frac{\partial}{\partial t}E_k\Big)\\ &= \frac{1}{2}\Big(E_j\varepsilon_{kj}\frac{\partial}{\partial t}E_k + E_j\varepsilon_{jk}\frac{\partial}{\partial t}E_k\Big).\end{aligned} \tag{1.6}$$

The equation is only valid if $\varepsilon_{jk} = \varepsilon_{kj}$ which means that $\varepsilon_{ij}(\omega)$ is symmetric.

1.2 Pockels effect

The electro-optical effetct induces birefringence in a crystal (for example KDP with a thickness of d). The resulting phase difference between an ordinary and an extraordinary wave is given by

$$\Delta\varphi = 2\pi n_0^3 r_{63} E \frac{d}{\lambda}. \tag{1.7}$$

1. *What voltage/unit length is necessary for a phase shift of π (KDP: $n_0 = 1.520; r_{63} = 23{,}3\,\frac{\mathrm{pm}}{\mathrm{V}}$? Assume a wavelength of 800 nm).*
2. *Estimate the capacity of the crystal (assume a cubic crystal with $d = 1\,\mathrm{cm}$) and the related charging time, if a $50\,\Omega$ resistor is used. (Relative permittivity of KDP $\varepsilon_r = 50$)*
3. *This effect can be used as a fast optical shutter. Estimate the cut-off (maximum) frequency of this shutter.*

a) Solution: For a phase shift of π which rotates the polarization by 90°, the Voltage $U = E \cdot d$ of the Pockels cell can be calculated by using (1.7).

$$U = E \cdot d = \frac{\lambda}{2 n_0^3 r_{63}} = \frac{800\,\mathrm{nm}}{2(1.520)^3 23{,}3\,\frac{\mathrm{pm}}{\mathrm{V}}} = 4888\,\mathrm{V}. \tag{1.8}$$

b) Solution: For the calculations we assume, that the cubic crystal is housed by two metal plates of same area. The capacity of a plate capacitor is given as

$$C = \varepsilon_0 \varepsilon_r \frac{A}{d} = \varepsilon_0 50 \frac{1\,\mathrm{cm}^2}{1\,\mathrm{cm}} = 4{,}43\,\mathrm{pF}. \tag{1.9}$$

The charging of the capacitor behaves like an exponential function of the following form:

$$Q(t) = U_0 \cdot C\left(1 - \exp\left(-\frac{t}{RC}\right)\right), \tag{1.10}$$

where RC represents the characteristic time for which the charge has risen to $(e-1)/e$ of its maximum value. Therefore the charging time is identified by this quantity:

$$\tau = R \cdot C = 50\,\Omega \cdot 4{,}43\,\mathrm{pF} = 221{,}5\,\mathrm{ps}. \tag{1.11}$$

Therefore the crystal/capacitor is charged in less than 1 ns.

c) Solution: Because the capacitor also has to discharge after turning off the voltage, the actual time it takes for a single cycle in the shutting process is about 1 ns. The maximum frequency of this shutter is therefore

$$f_{\mathrm{shutter}} = \frac{1}{1\,\mathrm{ns}} = 1\,\mathrm{GHz}. \tag{1.12}$$

However, the power requirement for a continuous operation of this shutter is actually quite high. The power can be calculated by the energy of the capacitor divided by the time of a single shutting cycle

$$P = \frac{Q \cdot U}{2\tilde{\tau}} = \frac{C \cdot U^2}{2\tau} \approx 53\,\mathrm{kW}. \tag{1.13}$$

This shows that this high power would heat up the resistor enormously. Therefore it is better to use lower frequencies of $f_{\mathrm{shutter}} = 1\,\mathrm{kHz}$, where the needed power drops to 0,053 W.

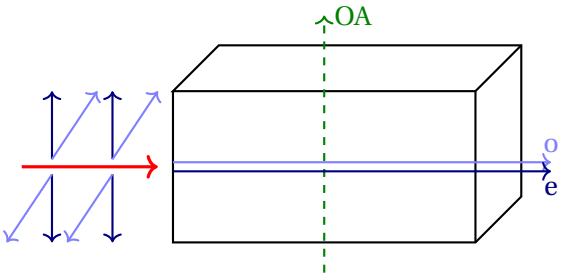
1.3 Birefringence

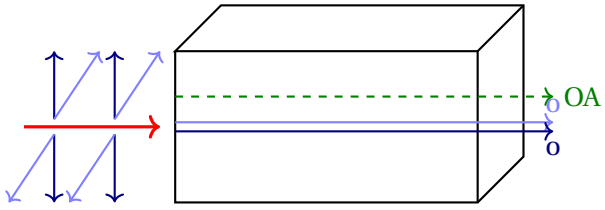
Answer the following questions for the 4 different orientations of a birefringent crystal (optic axis in green) depicted below:

- ***a)*** *If the incident light (red) is unpolarized, is it single or double refracted?*
- ***b)*** *Is there any phase retardation between different polarization states?*
- ***c)*** *Sketch the ordinary and extraordinary light rays inside the crystals!*
- ***d)*** *What happens in the 4 different cases if the incident light is linearly polarized in the plane of the sheet of paper?*
- ***e)*** *What happens in the 4 different cases if the incident light is linearly polarized perpendicular to the plane of the sheet of paper?*

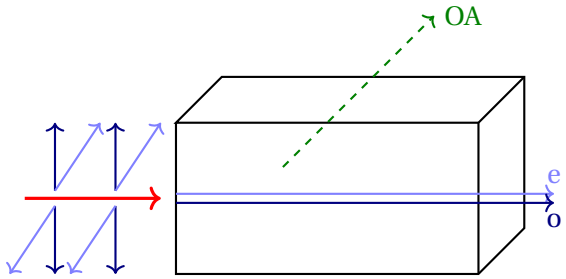
Solution

1.) For the first case, the incident unpolarized light is only single reflected, because for both polarization states (parallel or perpendicular to the optical axis) the polarization *sees* only one refractive index. However a phase retardation of the ordinary and extraordinary ray occurs. If the incident light is linearly polarized in the plane of the sheet of paper, the polarization is parallel to the optical axis and thus the extraordinary beam. For polarization perpendicular to the sheet of paper we get the ordinary beam. Both beams are not refracted, because they are normal to the crystal.

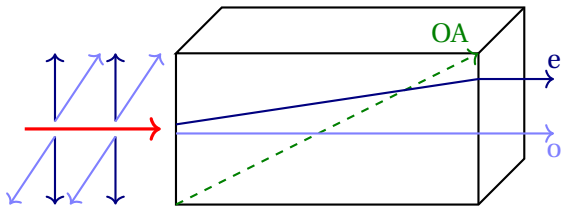




2.) For the second case, the incident unpolarized light is only single reflected, because all polarization states *see* only the ordinary refractive index. Every beam is an ordinary beam. Therefore no phase retardation occurs. Both polarization states (parallel and perpendicular to the plane of the sheet of paper) are ordinary beams. This special case only applies for propogation directions parallel to the optical axis.



3.) For the third case, the incident unpolarized we have the same scenario as in case 1. The only difference is, that the optical axis is now parallel to the polarization perpendicular to the plane of the sheet of paper. The ordinary beam becomes the extraordinary and vice versa. There is no double refraction occuring, however, phase retardation between ordinary and extraordinary beam still occurs.



4.) For the last case, there occurs double refraction for unpolarized light. The parts that are parallel polarized to the optical axis get refracted in the crystal, which leads to a separation of the two polarization states. There also occurs a phase retardation between the ordinary and the extraordinary beam, however, it is not as large as in case three. For a positive birefringence (the polarisation perpendicular to the optical axis is the fast ray) $n_e > n_o$ the extraordinary ray will be refracted to smaller angles with respect to the optical axis. Polarization the plane of the sheet of paper is the double refracted extraordinary ray, whereas the polarization perpendicular to the plane is the ordinary beam, which is not double reflected.

1.4 Half-wave plate

Birefringent crystals are widely used to change the polarization state of light beams. Crystalline quartz SiO_2, for example, can be used to change the polarization by exploiting the different refractive indices for ordinary and extraordinary axis.

- ***a)*** *Calcularte the phase retardation per millimetre for a linearly polarized Ti:Sa-Laser beam (800 nm), when the crystal is oriented to maximize the retardation!*
- ***b)*** *What crystal thickness is necessary to produce a half-wave-plate or a quarter-wave-plate?*

c) *It the incoming beam is linearly polarized at 30° with respect to the optical axis, calculate the angle at the output of the half-wave-plate!*

a.) Solution: The phase accumulated by the laser beam travelling through the medium can be expressed by $\varphi = k(n) \cdot L$. Then we can write the phase per length as

$$\frac{\varphi}{L} = k(n) = \frac{2\pi}{\lambda(n)} = \frac{2\pi n \nu}{c} = \frac{2\pi n}{\lambda_0}. \tag{1.14}$$

For crystalline quartz, the refractive indices for ordinary and extraodinary light are

$$n_o = 1.5383 \quad \text{and} \quad n_e = 1.5472 \qquad \Rightarrow \Delta n = 1.5383 - 1.5472 = 0.0089. \tag{1.15}$$

The phase difference between the ordinary and extraordinary beam can now be written as

$$\frac{\Delta\varphi}{L} = \frac{2\pi\Delta n}{\lambda_0} = \frac{2\pi \cdot 0.0089}{800\,\text{nm}} = 22.25\pi\,\frac{1}{\text{mm}}. \tag{1.16}$$

b.) Solution: For a half-wave plate the phase difference $\Delta\varphi$ must be equal to π. Therefore we can rewrite (1.16)

$$L = \frac{\lambda_0 \Delta\varphi}{2\pi\Delta n} = \frac{\lambda_0}{2\Delta n} = 45\,\mu\text{m}. \tag{1.17}$$

For a quarter wave plate the phase difference is $\Delta\varphi = \pi/2$ which leads to

$$L = \frac{\lambda_0 \Delta\varphi}{2\pi\Delta n} = \frac{\lambda_0}{4\Delta n} = 22{,}5\,\mu\text{m}. \tag{1.18}$$

c.) Solution: For a half-wave plate, the polarization is phase shifted by an angle of π. This leads to the change of the sign of one polarization direction. This can be interpreted as a reflection of the polarization vector at the optical axis. For an angle $\alpha = 30°$ the actual polarization rotation is therefore $2\alpha = 60°$. This corresponds to the trivial case of parallel or perpendicular polarization, for which no double refraction occurs.

2 Second Exercise

2.1 Estimate of nonlinearity coefficients

a) *Calculate the corresponding electric field strength necessary to bind the electron in a hydrogen atom! What intensity is required to reach such a field strength with a laser?*

b) *Use the Taylor expansion of the nonlinear polarization to estimate the order of magnitude for the $\chi^{(2)}$ and $\chi^{(3)}$ coefficients. Assume $\chi^{(1)} \approx 1$ (transparent solid media). The nonlinear terms in the expansion become relevant when the medium polarization approaches the atomic electric field strength.*

a.) Solution: The typical distance of the electron from the nucleus of the hydrogen atom is given by the Bohr radius

$$a_0 = \frac{4\pi\varepsilon_0\hbar^2}{m_e e^2} \approx 1\,\text{Å}. \tag{2.1}$$

The electric field strength of the proton at the Bohr radius is given by Coulombs law

$$E(r) = \frac{1}{4\pi\varepsilon_0}\frac{q}{a_0^2} \approx 5{,}76 \cdot 10^{11}\,\frac{\text{V}}{\text{m}}. \tag{2.2}$$

The intensity can be calculated as the temporal mean of the Poynting vector which results in

$$I = \frac{c\varepsilon_0}{2}|\boldsymbol{E}|^2 = 4{,}4 \cdot 10^{20}\,\frac{\text{W}}{\text{m}^2}. \tag{2.3}$$

b.) Solution: The Taylor expansion of the nonlinear polarization can be written as follows

$$\tilde{P}(t) = \varepsilon_0[\chi^{(1)}E(t) + \chi^{(2)}E^2(t) + \chi^{(3)}E^3(t) + \ldots]. \tag{2.4}$$

If we assume $\chi^{(1)} \approx 1$ the linear polarization term is of the order of $5.76 \cdot 10^{11}$. If the second order term is of the same magnitude as the linear polarization, then $\chi^{(2)}$ must be of the order of

$$\chi^{(2)} \sim \frac{1}{|E|} \sim 1.7 \cdot 10^{-12}. \tag{2.5}$$

For the third order nonlinearity term $\chi^{(3)}$ is of the order of

$$\chi^{(3)} \sim \frac{1}{|E|^2} \sim 3 \cdot 10^{-24}. \tag{2.6}$$

2.2 Focusing of high power beam

Imagine you have 1 kHz *repetition ultra-short pulse Ti:Sa-Laser* (800 nm) *system delivering* 1 mJ *pulses with a pulse duration of* 25 fs *(intensity FWHM).*

a) *What kind of lens do you need to focus the laser pulses to an intensity of* $10^{14}\,\frac{W}{cm^2}$ *assuming a Gaussian shaped pulse and beam (diameter* $d = 10\,\text{mm}$*, intensity drops to* $1/e^2$ *of its maximum)?*

b) *What will you do to achieve a peak intensity of* $10^{16}\,\frac{W}{cm^2}$*?*

Solution: We can calculate the pulse energy by integrating the intensity over the whole x-y-plane and time. We then get

$$E = \int_{-\infty}^{\infty} P(t)\,\mathrm{d}t = \int_{-\infty}^{\infty} \mathrm{d}t \int_0^{2\pi}\int_0^{\infty} r\,\mathrm{d}r\,\mathrm{d}\varphi\, I(t). \tag{2.7}$$

We can express the intensity of a Gaussian shaped pulse in the following way:

$$I(r,z) = I_0 \exp\left(-\eta\frac{r^2}{(2w_0')^2}\right)\cdot\exp\left(-\eta\frac{t^2}{\tau^2}\right), \tag{2.8}$$

where η is a variable which differs for various definitions of the pulse length and beam width

$$\eta = \begin{cases} 4\ln(2) & \text{FWHM} \\ 4 & 1/e \\ 8 & 1/e^2 \end{cases}. \tag{2.9}$$

For the criteria given in the task we get

$$I(r,z) = I_0 \exp\left(-2\frac{r^2}{(2w_0')^2}\right)\cdot\exp\left(-4\ln(2)\frac{t^2}{\tau^2}\right). \tag{2.10}$$

Then we can calculate the pulse energy to find an expression for the intensity I_0 in the beam waist

$$\begin{aligned} E &= \int_{-\infty}^{\infty} \exp\left(-4\ln(2)\frac{t^2}{\tau^2}\right)\mathrm{d}t \int_0^{2\pi}\int_0^{\infty} I_0 r \exp\left(-2\frac{r^2}{(2w_0')^2}\right)\mathrm{d}r\,\mathrm{d}\varphi \\ &= 2\pi\sqrt{\pi}\ln 2\frac{\tau}{2} I_0 \int_0^{\infty}\frac{1}{2}\exp\left(-\frac{2u}{w_0^2}\right)\mathrm{d}u \\ E &= \sqrt{\frac{\pi}{\ln 2}}\frac{\tau}{4} I_0 \pi w_0^2. \end{aligned} \tag{2.11}$$

Now we need to determine, how the diameter of the beam waist changes for a lens of focal length f. For that we assume, that we position the lens in the waist of the Gaussian beam. Then we can use the matrix algebra to determine the q-parameter in the new beam waist

$$\begin{pmatrix} 1 & f \\ 0 & 1 \end{pmatrix} \cdot \begin{pmatrix} 1 & 0 \\ -\frac{1}{f} & 1 \end{pmatrix} = \begin{pmatrix} 0 & f \\ -\frac{1}{f} & 1 \end{pmatrix} = \begin{pmatrix} A & B \\ C & D \end{pmatrix}. \tag{2.12}$$

The q-parameter is defined as

$$q = z - \mathrm{i} z_R \qquad \text{with } z_R = \frac{\pi w_0^2}{\lambda}, \tag{2.13}$$

where z is the distance to the beam waist and z_R the Rayleigh length. The new q-parameter for a given matrix can be calculated as

$$\begin{aligned} q_{\text{out}} &= \frac{A q_{\text{in}} + B}{C q_{\text{in}} + D}, \qquad q_{\text{in}} = \mathrm{i}\frac{\pi w_0^2}{\lambda} \\ &= \frac{f}{-\frac{q}{f} + 1} = \frac{f}{-\mathrm{i}\frac{\pi w_0^2}{\lambda f} + 1}. \end{aligned} \tag{2.14}$$

Now we can use the properties of the inverse q-parameter

$$\begin{aligned} \frac{1}{q} &= \frac{1}{R} + \mathrm{i}\frac{\lambda}{\pi}\frac{1}{w(z)^2} \\ \Rightarrow \frac{1}{q_{\text{out}}} &= \frac{1}{f} + \mathrm{i}\frac{\pi w_0^2}{\lambda f^2}. \end{aligned} \tag{2.15}$$

We can compare the imaginary parts to find that

$$\frac{\pi w_0^2}{\lambda f^2} = \frac{\lambda}{\pi}\frac{1}{w_0'^2} \quad \Rightarrow \quad \boxed{w_0' = \frac{\lambda f}{\pi w_0}}. \tag{2.16}$$

We can rearrange equation (2.11) to w_0' and compare it to (2.16)

$$w_0' = \sqrt{\frac{4E}{\tau \pi I_0}} \sqrt[4]{\frac{\ln(2)}{\pi}} \overset{!}{=} \frac{\lambda f}{\pi w_0}, \tag{2.17}$$

which leads to an expression for the focal length f

$$\begin{aligned} f &= \frac{\pi w_0}{\lambda} \sqrt{\frac{4E}{\tau \pi I_0}} \sqrt[4]{\frac{\ln(2)}{\pi}} \\ &= \frac{\pi \cdot 5\,\text{mm}}{800\,\text{nm}} \sqrt{\frac{4 \cdot 1\,\text{mJ}}{25\,\text{fs} \cdot \pi \cdot 10^{14}\,\frac{\text{W}}{\text{cm}^2}}} \sqrt[4]{\frac{\ln(2)}{\pi}} = 3{,}037\,\text{m}. \end{aligned} \tag{2.18}$$

For a desired peak intensity of $10^{16}\,\frac{\text{W}}{\text{cm}^2}$ the lens used to focus the beam must have a smaller focal length of

$$f = 30\,\text{cm}, \tag{2.19}$$

because the focal length is proportional to the inverse square root of the intensity.

2.3 Second order nonlinearities

The optical response of a medium can be described by developing the polarization $\tilde{P}(t)$ in a power series in the electric field strength $\tilde{E}(t)$ with pre-factors know as nonlinear susceptibilities $\chi^{(n)}$ as

$$\tilde{P}(t) = \tilde{P}^{(1)}(t) + \tilde{P}^{(2)}(t) + \tilde{P}^{(3)}(t) + \ldots = \varepsilon_0[\chi^{(1)}\tilde{E}(t) + \chi^{(2)}\tilde{E}^2(t) + \chi^{(3)}\tilde{E}^3(t) + \ldots]. \tag{2.20}$$

a) *Which type of material has an almost instantaneous response to the electric field such that equation (2.20) is valid? Briefly explain why.*

b) *Consider only $\chi^{(2)}$ processes. Derive an expression for second-order nonlinear interaction with two laser frequencies (ω_1 and ω_2). Name the related nonlinear phenomena.*

a.) Solution: Only materials without any losses and no dispersion have an almost instantaneous response to the electric field.

b.) Solution: We model the incident electric field as the sum of two fields with frequency ω_1 and ω_2

$$\tilde{E}(t) = E_1 \exp(-\mathrm{i}\omega_1 t) + E_2 \exp(-\mathrm{i}\omega_2 t). \tag{2.21}$$

We can now write the second order polarization as

$$\begin{aligned}
\tilde{P}^{(2)}(t) &= \varepsilon_0\chi^{(2)}\left[E_1\mathrm{e}^{-\mathrm{i}\omega_1 t} + E_2\mathrm{e}^{-\mathrm{i}\omega_2 t} + c.c.\right]^2 \\
&= \varepsilon_0\chi^{(2)}\left[E_1\mathrm{e}^{-\mathrm{i}\omega_1 t} + E_2\mathrm{e}^{-\mathrm{i}\omega_2 t} + E_1^*\mathrm{e}^{\mathrm{i}\omega_1 t} + E_2^*\mathrm{e}^{\mathrm{i}\omega_2 t}\right]^2 \\
&= \varepsilon_0\chi^{(2)}\big[(E_1\mathrm{e}^{-\mathrm{i}\omega_1 t} + E_2\mathrm{e}^{-\mathrm{i}\omega_2 t})^2 + 2(E_1\mathrm{e}^{-\mathrm{i}\omega_1 t} + E_2\mathrm{e}^{-\mathrm{i}\omega_2 t})(E_1^*\mathrm{e}^{\mathrm{i}\omega_1 t} + E_2^*\mathrm{e}^{\mathrm{i}\omega_2 t}) \\
&\qquad + (E_1^*\mathrm{e}^{\mathrm{i}\omega_1 t} + E_2^*\mathrm{e}^{\mathrm{i}\omega_2 t})^2\big] \\
&= \varepsilon_0\chi^{(2)}\big[E_1^2\mathrm{e}^{-\mathrm{i}2\omega_1 t} + E_2^2\mathrm{e}^{-\mathrm{i}2\omega_2 t} + 2E_1E_2\mathrm{e}^{-\mathrm{i}(\omega_1+\omega_2)t} + 2E_1E_1^* + 2E_2E_2^* \\
&= \qquad\qquad + 2E_1E_2 * \mathrm{e}^{-\mathrm{i}(\omega_1-\omega_2)t} + c.c.\big].
\end{aligned} \tag{2.22}$$

We can now identify the colors as Second Harmonic Generation (SHG), Sum frequency Generation (SFG), Optical Rectifictation (OR) and Difference Frequency Generation (DFG).

2.4 Supercontinuum generation

Supercontinuum generation can occur in media with Kerr nonlinearity, so all optical fields are coupled via the $\chi^{(3)}$ coefficient of the material. For the sake of simplicity, we only consider two electromagnetic waves (ω_1) and $(\omega_2) = \omega_1 + \Delta\omega$, spaced in frequency by $\Delta\omega$.

a) *If those fields interact in the Kerr medium, which new optical fields can be produced?*

b) *If these new generated optical fields have sufficiently high intensities, what will happen?*

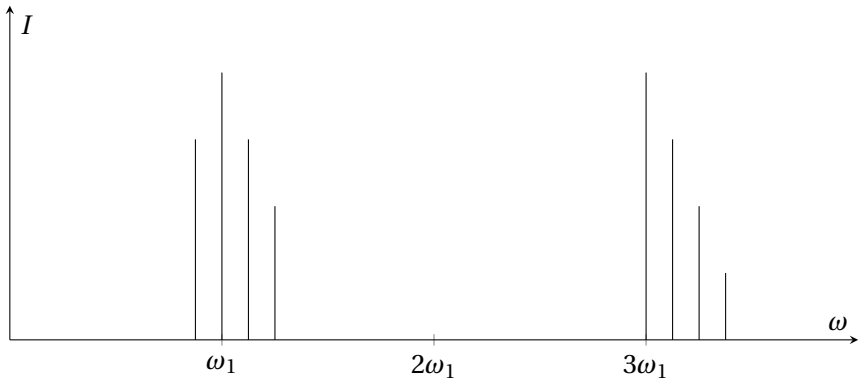
c) *Give a graphical representation of the spectrum for this process!*

a.) Solution: In order to assess, which optical fields can be produced, we calculate the third order nonlinear polarization term

$$\begin{aligned}
\tilde{P}^{(3)}(t) &= \varepsilon_0\chi^{(3)}\left[E_1 e^{-i\omega_1 t} + E_2 e^{-i(\omega_1+\Delta\omega)t} + c.c.\right]^3 \\
&= \varepsilon_0\chi^{(3)}\left[E_1 e^{-i\omega_1 t} + E_2 e^{-i(\omega_1+\Delta\omega)t} + E_1^* e^{i\omega_1 t} + E_2^* e^{i(\omega_1+\Delta\omega)t}\right]^3 \\
&= \varepsilon_0\chi^{(3)}\big[(E_1 e^{-i\omega_1 t} + E_2 e^{-i(\omega_1+\Delta\omega)t})^3 \\
&\qquad + 3(E_1 e^{-i\omega_1 t} + E_2 e^{-i(\omega_1+\Delta\omega)t})\cdot(E_1^* e^{i\omega_1 t} + E_2^* e^{i(\omega_1+\Delta\omega)t}) + c.c.\big] \\
&= \varepsilon_0\chi^{(3)}\big[E_1^2 e^{-i3\omega_1 t} + 3E_1^2 E_2 e^{-i(3\omega_1+\Delta\omega)t} + 3E_1 E_2^2 e^{-i(3\omega_1+2\Delta\omega)t} + 3E_2^3 e^{-i(3\omega_1+3\Delta\omega)t} \\
&\qquad + 3E_1^2 E_1^* e^{-i\omega_1 t} + 6E_1 E_2 E_1^* e^{-i(\omega_1+\Delta\omega)t} + 3E_2^2 E_1^* e^{-i(\omega_1+2\Delta\omega)t} \\
&\qquad + 3E_1^2 E_2^* e^{-i(\omega_1-\Delta\omega)t} + 6E_1 E_2 E_2^* e^{-i\omega_1 t} + 3E_2^2 E_2^* e^{-i(\omega_1+\Delta\omega)t} + c.c.\big] \qquad (2.23)
\end{aligned}$$

b-c.) Solution: For sufficiently high intensities the new generated optical fields can again create new frequencies by third order nonlinear frequency combination which leads to a very broad spectrum which is called a supercontinuum.

We can visualize the spectrum of the new optical fields we calculated in Task a)



3 Third Exercise

3.1 Difference Frequency Generation

Consider an optical parametric amplifier which consists of a lithium niobate crystal and a Nd:YAG pump laser (1064 nm):

- ***a)*** *If the amplifier is seeded with a weak optical field of $\lambda_2 = 1550\,\text{nm}$, what wavelengths are expected outside the crystal due to DFG? What are the corresponding photon energies for the pump, the seed and the output wave? Is the energy conservation fulfilled?*
- ***b)*** *Calculate the momenta of the involved photons for the DFG process (3 waves). If the pump and the seed are collinear into the crystal, is the momentum conservation fulfilled? Please interpret the results.*
- ***c)*** *For collinear case, calculate the accumulated linear phase shift through a crystal (thickness $d = 10\,\text{mm}$) for three waves. Compare the phase difference and interpret the results.*

The Sellmeier equation (which gives the real refractive index as a function of wavelength) for lithium niobate reads as (λ in μm) T in °C:

$$n^2 = a_1 + b_1 f + \frac{a_2 + b_2 f}{\lambda^2 - (a_3 + b_3 f)^2} + \frac{a_4 + b_4 f}{\lambda^2 - a_5^2} - a_6 \lambda^2 \tag{3.1}$$
$$f = (T - 24{,}5\,^\circ\text{C})(T + 570{,}82\,^\circ\text{C}).$$

$a_1 = 5.35583$, $a_2 = 0.100473$, $a_3 = 0.20692$, $a_4 = 100$, $a_5 = 11.34927$, $a_6 = 1.5334 \cdot 10^{-2}$
$b_1 = 4.629 \cdot 10^{-7}$, $b_2 = 3.862 \cdot 10^{-8}$, $b_3 = 0.89 \cdot 10^{-8}$, $b_4 = 2.657 \cdot 10^{-5}$.

a.) Solution: We can calculate the output wavelength by using the condition for DFG $\omega_3 = \omega_1 - \omega_2$, where the new generated frequency ω_3 is called the *idler* wave, whereas ω_2 is the *signal* wave. We therefore obtain

$$\omega_3 = \omega_1 - \omega_2 = 2\pi c\left(\frac{1}{\lambda_1} - \frac{1}{\lambda_2}\right) \quad \Rightarrow \quad \lambda_3 = 2\pi \frac{c}{\omega_3} = \frac{\lambda_1 \lambda_2}{\lambda_2 - \lambda_1} = 3{,}393\,\mu\text{m}. \tag{3.2}$$

We can calculate the photon energies by using $E_i = \hbar\omega_i$. We then obtain

$$E_1 = 1{,}165\,\text{eV} \qquad E_2 = 0{,}8\,\text{eV} \qquad E_3 = 0{,}3654\,\text{eV}. \tag{3.3}$$

The energy consveration is naturally fulfilled because

$$\hbar\omega_3 = \hbar(\omega_1 - \omega_2) \quad \Rightarrow \quad E_3 = E_1 - E_2. \tag{3.4}$$

b.) Solution: We can calculate the momentum via the wave vector $\boldsymbol{k}$

$$\boldsymbol{p} = \hbar \boldsymbol{k} \quad |\boldsymbol{k}| = \frac{2\pi\, n(\lambda)}{\lambda}. \tag{3.5}$$

We can calculate the refractive index for all wavelengths using (3.1) at 24,5 °C. This yields

$$n_1 = 2.1558 \qquad n_2 = 2.1379 \qquad n_3 = 2.0821. \tag{3.6}$$

We can now calculate the modulus of the momenta

$$p_1 = 1{,}3425 \cdot 10^{-27}\,\mathrm{J\,s} \qquad p_2 = 0{,}9139 \cdot 10^{-27}\,\mathrm{J\,s} \qquad p_3 = 0{,}4066 \cdot 10^{-27}\,\mathrm{J\,s}. \tag{3.7}$$

We can see that $p_1 - p_2 = 0{,}429 \cdot 10^{-27}\,\mathrm{J\,s} \neq p_3$. This means, that for the collinear case momentum conservation is not fulfilled. Therefore we cannot achieve DFG for the collinear case.

c.) Solution: We can calculate the accumulated phase by

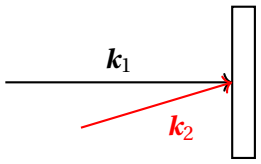
$$\varphi = k \cdot d = \frac{2\pi\, n}{\lambda} \cdot d. \tag{3.8}$$

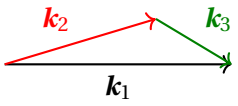
We can again compute all the phases for each wave which leads to

$$\varphi_1 = 127.305 \qquad \varphi_2 = 86662 \qquad \varphi_3 = 38558. \tag{3.9}$$

We can see that the phase difference is much larger than 2π, which means that the phases are not matched for the colinear case. This means, that the maxima of the electric fields do not overlap for the whole propagation through the crystal.

In order to obtain phase matching and momentum conservation we input the signal wave ω_2 under a small angle θ.





3.2 Coupled wave equations for DFG

In the lecture, the coupled wave equations for SFG ($\omega_3 = \omega_1 + \omega_2$) were derived using the nonlinear wave equation for all three frequencies

$$\Delta E_i - \frac{n_i^2}{c^2}\partial_t^2 E_i = \frac{1}{\varepsilon_0 c^2}\partial_t^2 P_i^{NL}. \tag{3.10}$$

a) *Derive the coupled wave equations for DFG ($\omega_2 = \omega_3 - \omega_1$).*

b) *By using the coupled wave equations, derive the Manley-Rowe relation for the process of DFG and give a brief discussion of the formulas.*

a.) Solution: For a small rhs of the nonlinear wave equations we can make the Ansatz

$$E_2(z,t) = A_2(z)\mathrm{e}^{\mathrm{i}(k_2 z - \omega_2 t)} + \text{c.c.} \tag{3.11}$$

The nonlinear source term can be written accordingly

$$P_2(z,t) = P_2\mathrm{e}^{-\mathrm{i}\omega_3 t} + \text{ c.c.} \tag{3.12}$$

We can write the spatial dependence of the polarization P_2 as

$$\begin{aligned} P_{\omega_2} = P_{\omega_3-\omega_1} = P_2 &= 4\varepsilon_0 d_{\text{eff}} E(\omega_3)E(-\omega_1) \\ &= 4\varepsilon_0 d_{\text{eff}} E(\omega_3)E^*(\omega_1) \\ &= 4\varepsilon_0 d_{\text{eff}} A_1^* A_3 \mathrm{e}^{\mathrm{i}(k_3-k_1)z}. \end{aligned} \tag{3.13}$$

Now we analyze all three terms of (3.10) separately:

$$\begin{aligned} \Delta E_2 &= \frac{\partial^2}{\partial z^2}\left[A_2(z)\mathrm{e}^{i(k_2 z-\omega_2 t)} + \text{c.c.}\right] \\ &= \frac{\partial}{\partial z}\left[A_2'(z)\mathrm{e}^{i(k_2 z-\omega_2 t)} + \mathrm{i}k_2 A_2 \mathrm{e}^{i(k_2 z-\omega_2 t)} + \text{c.c.}\right] \\ &= \left[\cancel{A_2''(z)} + \mathrm{i}k_2 A_2' - k_2^2 A_2 + \mathrm{i}k_2 A_2'\right]\mathrm{e}^{i(k_2 z-\omega_2 t)} + \text{c.c.} \\ &= \left[2\mathrm{i}k_2 A_2' - k_2^2 A_2\right]\mathrm{e}^{i(k_2 z-\omega_2 t)} + \text{c.c.} \end{aligned} \tag{3.14}$$

The second derivative of A_2 can be neglected, because we assume A_2 does not become very strong over short distances (Slowly varying amplitude approximation $\frac{\partial^2 A}{\partial z^2} \ll pdvAz$).
The second term can be evaluated as

$$\begin{aligned} -\frac{n_2^2}{c^2}\partial_t^2 E_2 &= \frac{n_2^2}{c^2}\omega_2^2 A_2(z)\mathrm{e}^{i(k_2 z-\omega_2 t)} + \text{c.c.} \\ &= k_2^2 A_2(z)\mathrm{e}^{i(k_2 z-\omega_2 t)} + \text{c.c.} \end{aligned} \tag{3.15}$$

Here we used the dispersion relation $k_2^2 = \frac{n_2^2}{c^2}\omega_2^2$.
The nonlinear term can be written as

$$\begin{aligned} \frac{1}{\varepsilon_0 c^2}\partial_t^2 P_i^{NL} &= \frac{1}{\varepsilon_0 c^2}\frac{\partial^2}{\partial t^2}\left[4\varepsilon_0 d_{\text{eff}} A_1^* A_3 \mathrm{e}^{\mathrm{i}(k_3-k_1)z-\omega_2 t} + \text{c.c.}\right] \\ &= \frac{-\omega_2^2}{\varepsilon_0 c^2}\left[4\varepsilon_0 d_{\text{eff}} A_1^* A_3 \mathrm{e}^{\mathrm{i}(k_3-k_1)z-\omega_2 t} + \text{c.c.}\right] \end{aligned} \tag{3.16}$$

Now we add (3.14), (3.15) and (3.16) together (neglect the complex conjugate part) and obtain

$$2\mathrm{i}k_2 A_2' \mathrm{e}^{\mathrm{i}k_2 z} = -\frac{-\omega_2^2}{\cancel{\varepsilon_0} c^2} 4\cancel{\varepsilon_0} d_{\mathrm{eff}} A_1^* A_3 \mathrm{e}^{\mathrm{i}(k_3-k_1)z}. \tag{3.17}$$

This result can be simplified by introducing the phase mismatch $\Delta k = k_3 - k_1 - k_2$

$$\frac{\partial A_2}{\partial z} = \mathrm{i}\frac{2\omega_2^2 d_{\mathrm{eff}}}{k_2 c^2} A_1^* A_3 \mathrm{e}^{\mathrm{i}\Delta k z}. \tag{3.18}$$

Analogously we can derive the coupled wave equations for A_1 and A_2

$$\frac{\partial A_1}{\partial z} = \mathrm{i}\frac{2\omega_1^2 d_{\mathrm{eff}}}{k_1 c^2} A_2^* A_3 \mathrm{e}^{\mathrm{i}\Delta k z} \tag{3.19}$$

$$\frac{\partial A_3}{\partial z} = \mathrm{i}\frac{2\omega_3^2 d_{\mathrm{eff}}}{k_3 c^2} A_1 A_2 \mathrm{e}^{-\mathrm{i}\Delta k z}. \tag{3.20}$$

b.) Solution: We can write the intensity I_i as

$$I_i = \frac{c n_i \varepsilon_0}{2} A_i A_i^*. \tag{3.21}$$

The derivative of the intensity is then

$$\begin{aligned}
\frac{\partial I_2}{\partial z} &= \frac{c n_i \varepsilon_0}{2}\left(A_2 \frac{\partial A_2 *}{\partial z} + A_2^* \frac{\partial A_2}{\partial z}\right) \\
&\overset{(3.18)}{=} \frac{c n_i \varepsilon_0}{2} \mathrm{i}\frac{2\omega_2^2 d_{\mathrm{eff}}}{k_2 c^2}\left(A_1^* A_2^* A_3 \mathrm{e}^{\mathrm{i}\Delta k z} + \mathrm{c.c.}\right) \\
&= \varepsilon_0 d_{\mathrm{eff}} \omega_2 \Bigg(\mathrm{i}\underbrace{A_1^* A_2^* A_3 \mathrm{e}^{\mathrm{i}\Delta k z}}_{\xi = a + \mathrm{i}b} + \mathrm{c.c.}\Bigg)
\end{aligned}$$

We can write the expression $\mathrm{i}\xi + \mathrm{c.c.} = \mathrm{i}(a+\mathrm{i}b) + (-\mathrm{i})(a-\mathrm{i}b) = -2b = -2\,\mathrm{Im}(\xi)$.

$$\frac{\partial I_2}{\partial z} = -2\varepsilon_0 d_{\mathrm{eff}} \omega_2 \,\mathrm{Im}(A_1^* A_2^* A_3 \mathrm{e}^{\mathrm{i}\Delta k z}) \tag{3.22}$$

$$\frac{\partial I_1}{\partial z} = -2\varepsilon_0 d_{\mathrm{eff}} \omega_1 \,\mathrm{Im}(A_1^* A_2^* A_3 \mathrm{e}^{\mathrm{i}\Delta k z}) \tag{3.23}$$

$$\frac{\partial I_3}{\partial z} = +2\varepsilon_0 d_{\mathrm{eff}} \omega_3 \,\mathrm{Im}(A_1^* A_2^* A_3 \mathrm{e}^{\mathrm{i}\Delta k z}). \tag{3.24}$$

If we now add up all results we obtain

$$\frac{\partial I_1}{\partial z} + \frac{\partial I_2}{\partial z} + \frac{\partial I_3}{\partial z} = 2\varepsilon_0 d_{\mathrm{eff}} \underbrace{(\omega_3 - \omega_1 - \omega_2)}_{=0} \mathrm{Im}(A_1^* A_2^* A_3 \mathrm{e}^{\mathrm{i}\Delta k z}) \tag{3.25}$$

which corresponds with energy conversation. We can now write down the MANLEY-ROWE relations for DFG

$$\boxed{\frac{\partial}{\partial z}\left(\frac{I_1}{\omega_1}\right) = \frac{\partial}{\partial z}\left(\frac{I_2}{\omega_2}\right) = -\frac{\partial}{\partial z}\left(\frac{I_3}{\omega_3}\right)}. \tag{3.26}$$

3.3 Perfect phase matching OPA

DFG can be used for optical parametric amplification (OPA) ($\omega_2 = \omega_3 - \omega_1$). *Assume a strong pump wave* ω_3 *which is undepleted* ($|A_3|$ *is constant). There is seed wave* (ω_2) *with small amplitude* $|A_2|$ *at the front surface of the crystal. The generated wave* ω_1 *from the DFG is called the idler wave* ($|A_1|$ *is* 0 *at the front surface).*

$$\frac{\mathrm{d}A_1}{\mathrm{d}z} = \frac{2\mathrm{i}\omega_1^2 d_{eff}}{k_1 c^2} A_3 A_2^* \mathrm{e}^{\mathrm{i}\Delta k z}, \quad \frac{\mathrm{d}A_2}{\mathrm{d}z} = \frac{2\mathrm{i}\omega_2^2 d_{eff}}{k_2 c^2} A_3 A_1^* \mathrm{e}^{\mathrm{i}\Delta k z} \tag{3.27}$$

with $\Delta k = k_3 - k_2 - k_1$. *Calculate the spatial evolution for* $|A_1(z)|^2$ *and* $|A_2(z)|^2$ *by using the coupled wave equation and give a graphical interpretation.*

Solution: For the undepleted pump approximation the amplitude of the wave ω_3 does not change $\frac{\mathrm{d}A_3}{\mathrm{d}z} = 0$. For the assumption of perfect phase matching $\Delta k = 0$ we can write the coupled wave equations as follows

$$\frac{\mathrm{d}A_1}{\mathrm{d}z} = \mu_1 A_3 A_2^*, \qquad \frac{\mathrm{d}A_2}{\mathrm{d}z} = \mu_2 A_3 A_1^*, \qquad \frac{\mathrm{d}A_3}{\mathrm{d}z} = 0, \qquad \mu_i = \mathrm{i}\frac{2\omega_1^2 d_{\mathrm{eff}}}{k_i c^2}. \tag{3.28}$$

We can decouple the system of differential equations by differentiation and substitution

$$\frac{\mathrm{d}^2 A_1}{\mathrm{d}z^2} = \mu_1 A_3 \frac{\mathrm{d}A_2^*}{\mathrm{d}z} = \mu_1 \mu_2^* A_3 A_3^* A_1 = \sigma^2 A_1 \qquad \sigma^2 = \mu_1 \mu_2^* |A_3|^2. \tag{3.29}$$

The simple solution for that differential equation is

$$A_1(z) = C_1(\mathrm{e}^{\sigma z} - \mathrm{e}^{-\sigma z}) = 2C_1 \sinh(\sigma z), \tag{3.30}$$

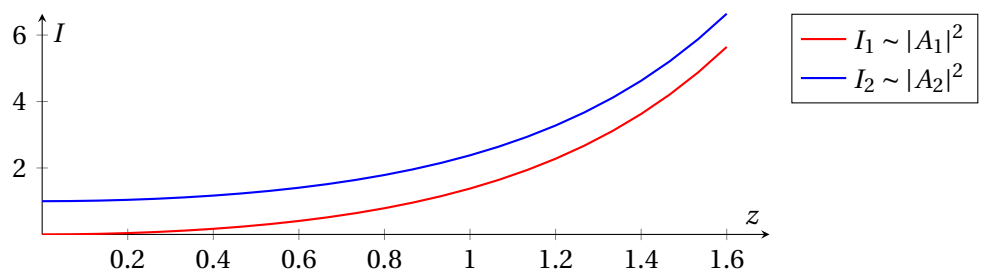
which fulfills the boundary condition $A_1(z) = 0$. We can use (3.28) to find $A_2(z)$

$$\begin{aligned} \frac{\mathrm{d}A_1}{\mathrm{d}z} &= \mu_1 A_3 A_2^* = C_1 \sigma (\mathrm{e}^{\sigma z} + \mathrm{e}^{-\sigma z}) \\ \Rightarrow A_2(z) &= \frac{\sigma C_1^*}{\mu_1^* A_3^*}(\mathrm{e}^{\sigma z} + \mathrm{e}^{-\sigma z}) + C_2 = \frac{2\sigma C_1^*}{\mu_1^* A_3^*} \cosh(\sigma z). \end{aligned} \tag{3.31}$$

We can calculate the intensity I of the two beams ω_1 and ω_2

$$I_1 \sim |A_1(z)|^2 = |A_1(0)|^2 \sinh^2(\sigma z) \tag{3.32}$$

$$I_2 \sim |A_2(z)|^2 = |A_2(0)|^2 \cosh^2(\sigma z). \tag{3.33}$$



4 Fourth Exercise

4.1 Phase Matching Bandwith of SH in BBO

Phase matching in Beta-Bariumborate (BBO) can be achieved by exploiting its birefringence similarly to KDP. You have a BBO crystal that is cut for efficient second harmonic generation (SHG) at 1,4 µm in collinear geometry (type 1 ooe phase matching, 90° to surface). Its thickness is $L = 50\,\mu\text{m}$.

a) *Make a sketch that indicates the polarization state of in-going and out-going radiation as well as the optic axis if you would mount the crystal in the lab to make SHG. Which axis needs to be aligned?*

b) *use a computer to calculate the wavelength dependent phase mismatch for the alignment described above for second harmonic generation in the wavelength range between 1,0 µm and 2,2 µm numerically. Plot the resulting* Δk *as function of the fundamental wavelength and indicate the phase matching bandwidth,* $\Delta\omega$*, where* $L\Delta k(\omega \pm \Delta\omega) \leq 2.7831$ *is satisfied. This corresponds to the half width of the conversion efficiency curve.*

c) *Estimate the phase matching bandwidth based on an analytical derivation to first order and compare it to your numerical result. Start from the definition of the phase mismatch, i. e.*

$$L\Delta k(\omega \pm \Delta\omega) \leq 2.7831, \tag{4.1}$$

where ω_0 *is the frequency of perfect phase matching. Write* Δk *into a Taylor series in order to obtain an expression for* $\Delta\omega$*. Afterwards, start from the expression for* Δk *in SHG and relate it to the refractive index of the material. Don´t forget to give the value for* $\Delta\omega$ *and mark* $\omega_0 \pm \Delta\omega$ *in the figure from b.) in the end.*

d) *Why is it important to consider the phase matching bandwidth if one wants to use the given crystal in a second harmonic generation auto-correlation?*

e) *Around which axis would you rotate the crystal if you want to make SHG from 1,8 µm with the same conversion efficiency as at 1,4 µm? You can mark it in the sketch from a.)*

a.) Solution: First we want to find the phase matching angle for the SHG at 1,4 µm. For that we try to find the refractive index $n_e(\lambda, \varphi)$ of the ordinary ray. We use the condition that the refractive index of the second harmonic for extraordinary rays must be equal to the refractive index of the fundamental:

$$n_e^2(\lambda/2, \varphi) = n_o^2(\lambda/2)\cos^2\varphi + n_e^2(\lambda/2)\sin^2\varphi \stackrel{!}{=} n_o^2(\lambda). \tag{4.2}$$

The analytic solution to find the angle is

$$\varphi = \pi - 2\arctan\left(\sqrt{\frac{\sqrt{(n_o^2 - n_e^2)(n_o^2 - n_e^2)}n_o^2 - 2n_e^2 + n_o^2}{n_o^2 - n_o^2}}\right). \tag{4.3}$$

We can calculate the refractive indices by using the Sellmeier-formula for BBO[1] which is given as

$$\begin{aligned} n_o^2(\lambda) &= 2.7359 + \frac{0.01878}{\lambda^2 - 0.01822} - 0.01354\lambda^2 \\ n_e^2(\lambda) &= 2.3753 + \frac{0.01224}{\lambda^2 - 0.01667} - 0.01516\lambda^2 \end{aligned}. \tag{4.4}$$

Using Python we can evaluate $n_e^2(0{,}7\,\mu\text{m}), n_o^2(0{,}7\,\mu\text{m}), n_o^2(1{,}4\,\mu\text{m})$ and find, that the angle ϕ is

$$\varphi = 0.3737665614033969 \approx 21{,}41^\circ. \tag{4.5}$$

We now cut the crystal in such a way, that the propagation axis is tilted to the optical axis by the angle φ. This is sketched in the figure 1:

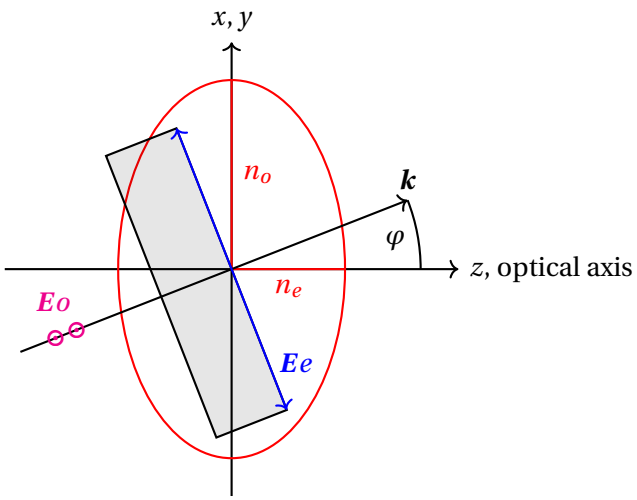


Fig. 1: Polarization state of in-going (fundamental) and out-goin (SH) radiation.

b.) Solution: We can calculate the wavelength dependent phase mismatch using the following formula:

$$\begin{aligned} \Delta k &= 2k_1 - k_2 = \frac{2\pi}{\lambda_0}(2n_1 - n_2) \\ &= 4\pi\big(n_o(\lambda) - n_e(\lambda/2, \varphi)\big). \end{aligned} \tag{4.6}$$

This can be plotted using Python. The graph is displayed in figure 2. We can calculate the

[1] https://de.wikipedia.org/wiki/Bariumborat

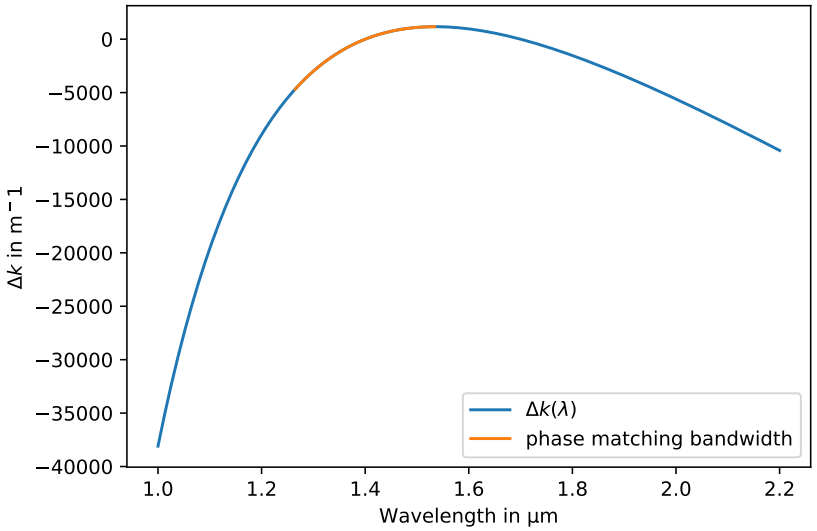


Fig. 2: Phase mismatch for BBO for SHG at 1,4 µm.

phase matching bandwidth by using

$$\Delta k(\omega_0 + \Delta\omega) \leq \frac{2.781}{50\,\mu\text{m}} = 55\,662\,\frac{1}{\text{m}}. \tag{4.7}$$

Graphically we can find the value $\Delta\lambda = 0{,}135\,\mu\text{m}$. We can relate this to the bandwidth $\Delta\omega$

$$\begin{aligned}\Delta\omega &= 2\pi c\left(\frac{1}{\lambda_1} - \frac{1}{\lambda_2}\right) = 2\pi c\left(\frac{\Delta\lambda}{\lambda_1 \cdot \lambda_2}\right) \\ &= 2\pi c\left(\frac{0{,}135\,\mu\text{m}}{1{,}4\,\mu\text{m}1{,}535\,\mu\text{m}}\right) = 118{,}3\,\text{THz}.\end{aligned} \tag{4.8}$$

d.) Solution: If we want to use the crystal in an autocorrelator we want to correlate the short pulse with itself. This requires phase matching over the whole bandwidth of the pulse, which can be, depending on the pulse length, rather broad. We therefore need a phase matching bandwidth that is larger than the bandwidth of the pulse.

e.) Solution: In order to achieve phase matching at another wavelength we need to adjust the angle φ. We can run the simulation again for $\lambda = 1{,}8\,\mu\text{m}$ which yields

$$\varphi' = 21{,}74°, \tag{4.9}$$

which means that we have to rotate around the plane of the wave vector $\boldsymbol{k}$ and the optical axis.

5 Fifth Exercise

5.1 Kleinman's symmetry and $\chi^{(2)}$ tensor

Quite often incident optical waves are far away from a resonance of the optical material. In that case the dispersion is negligible and the susceptibility does not depend on the frequencies. Then the indices can be permuted without permuting the frequencies. That is known as Kleinman's symmetry. Very often a tensor

$$d_{ijk} = \frac{1}{2}\chi^{(2)}_{ijk} \tag{5.1}$$

is defined without frequency arguments. For symmetric tensors the contracted notation d_{il} was introduced in the lecture. How many elements of d_{il} are independent when Kleinman's symmetry is valid? Show that by writing down the matrix d_{il}.

Solution: For a medium with Kleinman's symmetry we observe full permutation symmetry of the indices of χ_{ijk} meaning

$$d_{ijk} = d_{ikj} = d_{jik} = d_{jki} = d_{kij} = d_{kji}. \tag{5.2}$$

Since the susceptibility is a symmetric tensor we can introduce the contracted notation:

$$d_{ijk} \rightarrow d_{il} \quad \text{with} \quad \begin{matrix} d_{ijk} & 11 & 22 & 33 & 23 & 13 & 12 \\ d_{il} & 1 & 2 & 3 & 4 & 5 & 6 \end{matrix}. \tag{5.3}$$

In contracted notation the d_{il} tensor has 18 components and looks like

$$d_{il} = \begin{pmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{pmatrix}. \tag{5.4}$$

We can now write the d_{ij} tensor in the uncontracted components

$$d_{ijk} = \begin{pmatrix} d_{111} & d_{122} & d_{133} & d_{123} & d_{113} & d_{112} \\ d_{211} & d_{222} & d_{233} & d_{223} & d_{213} & d_{212} \\ d_{311} & d_{322} & d_{333} & d_{323} & d_{313} & d_{312} \end{pmatrix}. \tag{5.5}$$

We can see now that the following components of the d_{il} tensor are equal for Kleinman's symmetry:

$$\begin{aligned} &d_{12} = d_{26} && d_{13} = d_{35} && d_{16} = d_{21} \\ &d_{15} = d_{31} && d_{23} = d_{34} && d_{24} = d_{32} \\ &d_{14} = d_{25} = d_{36}. \end{aligned} \tag{5.6}$$

Therefore we have 10 independent components for Kleinman's symmetry.

5.2 Structure of d_{ijk} for symmetry class $3m$

Crystal symmetry simplifies the structure of the truncated tensor d_{il} even more. Consider a trigonal crystal system and the $3m$ crystal class (c_{3v} in Schönflies notation). c_n indicates that the group has a n-fold rotation axis (c means "cyclic"). c_{nv} has an additional mirror plane parallel to the axis of rotation.

Use the crystal symmetry to retrieve the form of the matrix d_{il}. Assume that the Kleinman's symmetry is valid.

Solution: The crystal class $3m$ is a *ditrigonal-pyramidial*. We assume that the axis of rotation is the x-axis. Therefore the mirror plane is the xy-plane.

We now need to write down the matrices for the symmetry transformations. First we consider the mirror symmetry of the xy-plane. This symmetry operation flips the sign of the z-coordinate. Therefore the corresponding matrix is

$$M = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -1 \end{pmatrix}. \tag{5.7}$$

Using Einsteins sum convention we can write the transformation of d_{ijk} as

$$d_{ijk} \stackrel{!}{=} d'_{ijk} = M_{il} M_{jm} M_{kn} d_{lmn}. \tag{5.8}$$

We need to apply M three times for each. Because M_{ij} is a diagonal matrix we find

$$\begin{aligned} d_{ijk} &= M_{ii} M_{jj} M_{kk} d_{lmn} \\ &= \begin{cases} +d_{ijk} & \text{for zero or two 3's} \\ -d_{ijk} & \text{for one or three 3's} \end{cases}. \end{aligned} \tag{5.9}$$

For the second case we have $d_{ijk} = -d_{ijk}$ which directly implies $d_{ijk} = 0$. So lets write down the elements of d_{ijk}:

$$\begin{array}{ccc|ccc|ccc} 111 & 112 & \cancel{113} & 211 & 212 & \cancel{213} & \cancel{311} & \cancel{312} & 313 \\ 121 & 122 & \cancel{123} & 221 & 222 & \cancel{223} & \cancel{321} & \cancel{322} & 323 \\ \cancel{131} & \cancel{132} & 133 & \cancel{231} & \cancel{232} & 233 & 331 & 332 & \cancel{333} \end{array} \tag{5.10}$$

Now we can see how the tensor d_{il} will look like:

$$d_{il} = \begin{pmatrix} d_{11} & d_{12} & d_{13} & 0 & 0 & d_{21} \\ d_{21} & d_{22} & d_{23} & 0 & 0 & d_{12} \\ 0 & 0 & 0 & d_{23} & d_{13} & 0 \end{pmatrix}. \tag{5.11}$$

Next we write down the Matrix for the 120° rotation around the x-axis. Here we simply take a rotational Matrix

$$R = \begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos\varphi & \sin\varphi \\ 0 & -\sin\varphi & \cos\varphi \end{pmatrix} \quad \text{with} \quad \varphi = 120^\circ. \tag{5.12}$$

We note that $\sin\big(\varphi(120°)\big) = \frac{\sqrt{3}}{2}$ and $\cos\big(\varphi(120°)\big) = -\frac{1}{2}$. So now lets look at the six indices in (5.11) in the upper left corner. In general d_{ijk} transforms as

$$d_{ijk} = R_{il}R_{jm}R_{kn}d_{lmn}. \tag{5.13}$$

For d_{111} this is simply

$$d_{111} = R_{1l}R_{1m}R_{1n}d_{lmn} = R_{11}R_{11}R_{11}d_{111} = d_{111}. \tag{5.14}$$

This is a trivial solution. Next we consider d_{212}

$$\begin{aligned} d_{222} &= R_{2l}R_{2m}R_{2n}d_{lmn} \\ &= \cos\varphi\cos\varphi\cos\varphi\, d_{222} \\ &\quad + (\sin\varphi\cos\varphi\cos\varphi)(\cancel{d_{322}} + \cancel{d_{232}} + \cancel{d_{223}}) \\ &\quad + \sin\varphi\sin\varphi\cos\varphi(d_{233}, d_{323}, d_{332}) \\ &\quad + \sin\varphi\sin\varphi\sin\varphi\,\cancel{d_{333}} \\ \Rightarrow d_{22} &= \cos^3\varphi d_{22} + 3\sin^2\varphi\cos\varphi d_{23} \\ d_{22} &= -\frac{1}{8}d_{22} - \frac{9}{8}d_{23} \Rightarrow d_{22} = -d_{23}. \end{aligned} \tag{5.15}$$

Analogously we can calculate d_{211}

$$\begin{aligned} d_{211} &= R_{2l}R_{1m}R_{1n}d_{lmn} \\ &= R_{22}R_{11}R_{11}d_{211} + R_{23}R_{11}R_{11}\cancel{d_{311}} \\ d_{211} &= \cos\varphi\, d_{211} \Rightarrow d_{21} = 0. \end{aligned} \tag{5.16}$$

and also d_{122}

$$\begin{aligned} d_{122} &= R_{1l}R_{2m}R_{2n}d_{lmn} \\ &= R_{11}R_{22}R_{22}d_{122} + R_{11}R_{22}R_{23}\cancel{d_{123}} + \cancel{d_{132}} + R_{11}R_{23}R_{23}d_{133} \\ &= \cos^2\varphi\, d_{122} + \sin^2\varphi\, d_{133} \\ \Rightarrow d_{122} &= d_{133} \Rightarrow d_{12} = d_{13}. \end{aligned} \tag{5.17}$$

Inserting these results into (5.11) we obtain

$$d_{il} = \begin{pmatrix} d_{11} & d_{12} & d_{12} & 0 & 0 & 0 \\ 0 & d_{22} & -d_{22} & 0 & 0 & d_{12} \\ 0 & 0 & 0 & -d_{22} & d_{12} & 0 \end{pmatrix}. \tag{5.18}$$

Therefore we can see, that the d_{il} tensor has only three independent indices.

6 Sixth Exercise

6.1 Optical Kerr Lens

An optical beam travelling in the z-direction is transmitted through a thin nonlinear optical material exhibiting Optical Kerr effect, $n(I) = n_0 + n_2 I$. The material lies in the xy-plane and has a small thickness d so that its complex amplitude transmittance is $\exp(-\mathrm{i}nk_0 d)$. *The beam has an approximately planar wavefront and an intensity distribution $I \approx I_0(1-(x^2+y^2)/w^2)$ at points near the beam axis ($x, y \ll w$), where I_0 is the peak intensity and w is the beam width.*

a) *Show that the medium acts as a thin lens with focal length f that is inversely proportional to I_0.*

b) *A collimated Gaussian ($w = 6\,\mathrm{mm}$) beam is focused into a vacuum chamber using a $f = 1\,\mathrm{m}$ lens. 10 cm after the focusing lens, the beam enters the vacuum chamber through a fused silica window that has a thickness of 2 mm. Use your results from a) to calculate the shift of the focus position due to the Kerr Lens that is formed in the entrance window at an intensity of $I_{KL} = 3\cdot 10^{11}\,\frac{\mathrm{W}}{\mathrm{cm}}$ ($n_2 = 3\cdot 10^{-16}\,\frac{\mathrm{cm}}{\mathrm{W}}$).*

Hint: *A lens of focal length f has a complex amplitude of transmittance proportional to* $\exp\left(\mathrm{i}k_0\frac{x^2+y^2}{2f}\right)$.

a.) Solution: In order to show that this nonlinear optical material acts as a lens, we proof that the complex amplitude of transmittance is the same as for a lens. We can write the electric field behind the material with thickness d as

$$\begin{aligned} E(x,y,z=d) &= E(x,y,z=0)\exp(-\mathrm{i}nk_0 d) \\ &= E(x,y,0)\exp(-\mathrm{i}(n_0+n_2 I)k_0 d) \\ &= E_0\exp\left(-\mathrm{i}\left(n_0+n_2 I_0\left(1-\frac{x^2+y^2}{w^2}\right)\right)k_0 d\right) \\ &= E_0\underbrace{\exp(-\mathrm{i}(n_0+n_2 I_0)k_0 d)}_{\text{const. phase}}\underbrace{\exp\left(-\mathrm{i}n_2 I_0\left(-\frac{x^2+y^2}{w^2}\right)k_0 d\right)}_{\text{evolving phase}}. \end{aligned} \tag{6.1}$$

We now compare the evolving phase to the phase transmittance of a lens

$$\begin{aligned} &\exp\left(-\mathrm{i}n_2 I_0\left(-\frac{x^2+y^2}{w^2}\right)k_0 d\right) \stackrel{!}{=} \exp\left(\mathrm{i}k_0\frac{x^2+y^2}{2f}\right) \\ &\Rightarrow \frac{n_2 I_0 k_0 d}{w^2} = \frac{k_0}{2f} \quad \Rightarrow \quad f = \frac{w^2}{2n_2 I_0 d}. \end{aligned} \tag{6.2}$$

We see that the self-focusing effect is larger for a longer medium d and higher intensities.

b.) Solution: Since we have a Gaussian beam we can use q-parameters to describe the beam evolution. We start with the inverse of the q-parameter

$$\frac{1}{q_0} = \frac{1}{R} - \mathrm{i}\frac{\lambda}{\pi w_0^2} = -\mathrm{i}\frac{\lambda}{\pi w_0^2} \quad \Rightarrow q_0 = \mathrm{i}\frac{\pi w_0^2}{\lambda} = \mathrm{i}z_R, \tag{6.3}$$

where z_R is the Rayleigh length.

We can now use the ABCD-matrix formalism to describe the behaviour of the q-parameter. Our goal is to find the beam width at the entrance of the nonlinear medium. The transfer matrix for a lens $f = 1\,\mathrm{m}$ and free space propagation $L = 0{,}1\,\mathrm{m}$ is

$$\begin{pmatrix} A & B \\ C & D \end{pmatrix} = \begin{pmatrix} 1 & L \\ 0 & 1 \end{pmatrix}\begin{pmatrix} 1 & 0 \\ -\frac{1}{f} & 1 \end{pmatrix} = \begin{pmatrix} 1-\frac{L}{f} & L \\ -\frac{1}{f} & 1 \end{pmatrix}. \tag{6.4}$$

The q-parameter can be written as

$$\frac{1}{q} = \frac{Cq_0 + D}{Aq_0 + B} = \frac{1 - \mathrm{i}\dfrac{z_R}{f}}{\mathrm{i}(1-\frac{L}{f})z_R + L} = \frac{1 - \mathrm{i}z_R}{0.1 + \mathrm{i}0.9z_R}. \tag{6.5}$$

We obtain the new beam radius by looking at the imaginary part of (6.5)

$$\begin{aligned} -\frac{\lambda}{\pi w^2} \overset{!}{=} \mathrm{Im}\left(\frac{1-\mathrm{i}z_R}{0.1+\mathrm{i}0.9z_R}\right) &= \mathrm{Im}\left(\frac{(1-\mathrm{i}z_R)(0.1-0.9\mathrm{i}z_R)}{0.1^2 + (0.9z_R)^2}\right) \\ -\frac{\lambda}{\pi w^2} &= -\frac{0.1z_R + 0.9z_R}{0.01 + 0.81z_R^2} \\ \frac{w_0^2}{z_R w^2} &= \frac{z_R}{0{,}01\,\mathrm{m}^2 + 0.81z_R^2} \\ \Rightarrow w^2 &= w_0^2\left(\frac{0{,}01\,\mathrm{m}^2}{z_R^2} + 0.81\right) \\ &= w_0^2\left(\frac{0{,}01\,\mathrm{m}^2\cancel{\lambda^2}}{\cancel{\pi^2} w_0^4} + 0.81\right) = 0.81\,w_0^2. \end{aligned} \tag{6.6}$$

We therefore conclude that the width of the beam has been reduced to $w = 0.9w_0 = 5{,}4\,\mathrm{mm}$. Now we can calculate the focal length of the nonlinear material by using (6.2)

$$f' = \frac{w^2}{2n_2 I_0 d} = \frac{(5{,}4\,\mathrm{mm})^2}{2\cdot 3\cdot 10^{-16}\,\frac{\mathrm{cm}}{\mathrm{W}} 3\cdot 10^{11}\,\frac{\mathrm{W}}{\mathrm{cm}}\cdot 2\,\mathrm{mm}} = 81\,\mathrm{m}. \tag{6.7}$$

We can calculate the new focal length of the system using

$$\begin{aligned} \frac{1}{f_{\mathrm{tot.}}} &= \frac{1}{f} + \frac{1}{f'} - \frac{L}{f_1 f_2} \\ &= 1\,\frac{1}{\mathrm{m}} + \frac{1}{80\,\mathrm{m}} - \frac{10\,\mathrm{cm}}{80\,\mathrm{m}^2} = 1{,}011\,25\,\frac{1}{\mathrm{m}} \\ \Rightarrow f_{\mathrm{tot}} &= 98{,}89\,\mathrm{cm}. \end{aligned} \tag{6.8}$$

The Focal position is shifted by about 1,11 cm.

6.2 Self-Phase Modulation

Consider a Gaussian shaped pulse which has a time-dependent intensity distribution: $I(t) = I_0 \exp\left(-(t/\tau)^2\right)$. *Assume the pulse is propagating along* z*-direction through a nonlinear optical material exhibiting strong optical Kerr effect,* $n(I) = n_0 + n_2 I$.

a) *Show that the instantaneous phase of the beam inside the martial has an intensity dependence and calculate the nonlinear phase shift due to the Kerr effect. (Considering a plane wave,* $E = E_0 \exp(-\mathrm{i}(\omega_0 t - kz))$*).*

b) *How does this nonlinear phase shift contribute to the frequency of the field and what can we use this for?*

a.) Solution: We can write the phase of a Gaussian pulse as

$$\phi(t) = \omega_0 t - kz = \omega_0 t - k_0 n z. \tag{6.9}$$

Now we use that the refractive index is intensity dependent. The phase will now transform as

$$\begin{aligned}\phi(t) &= \omega_0 t - k_0\left(n_0 + n_2 I_0 \exp\left(-\frac{t^2}{\tau^2}\right)\right)z \\ &= \underbrace{\omega_0 t - k_0 n_0 z}_{\text{linear phase}} \underbrace{- k_0 n_2 I_0 \exp\left(-\frac{t^2}{\tau^2}\right)z}_{\text{nonlinear phase}}.\end{aligned} \tag{6.10}$$

The instantaneous frequency is the time derivative of the temporal phase

$$\begin{aligned}\omega_{\text{inst.}} &= \frac{\mathrm{d}\phi_0}{\mathrm{d}t} + \frac{\mathrm{d}\Phi_{\text{NL}}}{\mathrm{d}t} = \omega_0 + \Delta\omega \\ &= \omega_0 - k_0 n_2 z \frac{\mathrm{d}I}{\mathrm{d}t}.\end{aligned} \tag{6.11}$$

We can calculate the phase shift $\Delta\omega$ as

$$\Delta\omega = k_0 n_2 z \frac{2t}{\tau^2} I(t). \tag{6.12}$$

b.) Solution: For the leading edge of the pulse ($t < 0$) the frequency shift is negative and for trailing edge ($t > 0$) positive. This corresponds to a positive frequency chirp imprinted on the pulse. In general, many new frequencies are generated by SPM which increases the bandwidth of the pulse by a considerable amount. The can use this effect to further reduce the pulse duration of a femtosecond laser pulse, where the pulse duration is limited by the gain narrowing effect which reduces the bandwidth and hence increases pulse duration.

Typically, a 25 fs pulse is focused into a hollow core fibre filled with a noble gas. Due to the SPM in the core the bandwidth of the pulse increases and hence reduces the Fourier limited pulse duration. The positive chirp is compensated by using multi-layered chirped mirrors.

6.3 Estimation of NL phase contributions B-integral

In order to estimate the nonlinear phase shift on an optical path one can use the so-called B-integral which is accumulated in the last pass of a high power Ti:Sa-Laser amplifier assuming a stretched pulse duration of 200 ps, a pulse energy of 13 mJ, a crystal thickness of 8 mm and a beam diameter of 300 µm. Please interpret the results.

Solution: The B-integral (beam breakup integral) is used to measure phase distortions inside the Kerr-medium of length l and is defined as

$$B = \frac{2\pi}{\lambda} \int_0^l n_2 I(z)\,\mathrm{d}z. \tag{6.13}$$

It describes the total nonlinear phase shift accumulated over the length of the crystal and therefore characterizes the importance of self-focusing effects.

We assume that the intensity of the laser pulse is constant on the beam axis but varies with time. For a Gaussian temporal shape the intensity can be written as

$$I(x,y,z,t) = I_0 \mathrm{e}^{-4\ln 2 \frac{t^2}{\tau^2}} \mathrm{e}^{-\frac{x^2+y^2}{w^2}}. \tag{6.14}$$

We obtain the total pulse energy by integrating over the xy-plane and time

$$\begin{aligned} E &= \int_{-\infty}^{\infty} \mathrm{d}t \int_0^{2\pi} \int_0^{\infty} r^2\,\mathrm{d}r\,\mathrm{d}\varphi\, I_0 \mathrm{e}^{-4\ln 2 \frac{t^2}{\tau^2}} \mathrm{e}^{-\frac{x^2+y^2}{w^2}} \\ &= \frac{2\pi I_0}{2} \int_{-\infty}^{\infty} \mathrm{d}t \int_0^{\infty} \mathrm{d}u\, \mathrm{e}^{-4\ln 2 \frac{t^2}{\tau^2}} \mathrm{e}^{-\frac{u}{w^2}} \\ &= \pi w^2 I_0 \int_{-\infty}^{\infty} \mathrm{d}t\, \mathrm{e}^{-4\ln 2 \frac{t^2}{\tau^2}} = \sqrt{\frac{\pi}{4\ln 2}} \pi w^2 I_0 \tau \approx 1.06 \pi w^2 I_0 \tau. \end{aligned} \tag{6.15}$$

We therefore find $I(z) = \frac{0.94E}{\pi w^2 \tau} = 8{,}644 \cdot 10^{10}\,\frac{\mathrm{W}}{\mathrm{cm}^2}$. Using $n_2 \approx 3 \cdot 10^{-16}\,\frac{\mathrm{cm}^2}{\mathrm{W}}$ we can use this result to calculate the B-integral

$$B = \frac{2\pi}{\lambda} d I_0 n_2 = \frac{2\pi}{800\,\mathrm{nm}} 8\,\mathrm{mm} \cdot 8{,}644 \cdot 10^{10}\,\frac{\mathrm{W}}{\mathrm{cm}^2} \cdot 3 \cdot 10^{-16}\,\frac{\mathrm{cm}^2}{\mathrm{W}} = 1.633 < 3. \tag{6.16}$$

For values of B which are smaller than three, the nonlinear effects do not play an important role.