LECTURE 9

In this lecture, we will focus on examples of electromagnetic wave propagation in nonlinear optical media, by applying the forms of Maxwell's equations as obtained in the eighth lecture to a set of particular nonlinear interactions as described by the previously formulated nonlinear susceptibility formalism.

The outline for this lecture is:
- General process for solving problems in nonlinear optics
- Second harmonic generation (SHG)
- Optical Kerr-effect

General process for solving problems in nonlinear optics

The typical steps in the process of solving a theoretical problem in nonlinear optics typically involve:

$$\left\{\begin{array}{c}\text{define the optical interaction of interest}\\ \text{(identifying the susceptibility)}\end{array}\right\}$$

$$\Downarrow$$

$$\left\{\begin{array}{c}\text{define in which medium the interaction take place}\\ \text{(identify crystallographic point symmetry group)}\end{array}\right\}$$

$$\Downarrow$$

$$\left\{\begin{array}{c}\text{consider eventual additional symmetries and constraints}\\ \text{(e. g. intrinsic, overall, or Kleinman symmetries)}\end{array}\right\}$$

$$\Downarrow$$

$$\left\{\begin{array}{c}\text{construct the polarization density}\\ \text{(the Butcher and Cotter convention)}\end{array}\right\}$$

$$\Downarrow$$

$$\left\{\begin{array}{c}\text{formulate the proper wave equation}\\ \text{(e. g. taking dispersion or diffraction into account)}\end{array}\right\}$$

$$\Downarrow$$

$$\left\{\begin{array}{c}\text{formulate the proper boundary}\\ \text{conditions for the wave equation}\end{array}\right\}$$

$$\Downarrow$$

$$\left\{\begin{array}{c}\text{solve the wave equation under}\\ \text{the boundary conditions}\end{array}\right\}$$

Formulation of the exercises in this lecture

In order to illustrate the scheme as previously outlined, the following exercises serve as to give the connection between the susceptibilities, as extensively analysed from a quantum-mechanical basis in earlier lectures of this course, and the wave equation, derived from Maxwell's equations of motion for electromagnetic fields.

Exercise 1. (*Second harmonic generation in negative uniaxial media*) Consider a continuous pump wave at angular frequency ω, initially polarized in the y-direction and propagating in the positive x-direction of a negative uniaxial crystal of crystallographic point symmetry group $3m$. (Examples of crystals belonging to this class: beta-BaB_2O_4/BBO, $LiNbO_3$.)

1a. Formulate the polarization density of the medium for the pump and second harmonic wave.

1b. Formulate the system of equations of motion for the electromagnetic fields.

1c. Assuming no second harmonic signal present at the input, solve the equations of motion for the second harmonic field, using the non-depleted pump approximation, and derive an expression for the conversion efficiency of the second harmonic generation.

Exercise 2. (*Optical Kerr-effect – continuous wave case*) In this setup, a monochromatic optical wave is propagating in the positive z-direction of an isotropic optical Kerr-medium.

2a. Formulate the polarization density of the medium for a wave polarized in the xy-plane.

2b. Formulate the polarization density of the medium for a wave polarized in the x-direction.

2c. Formulate the wave equation for continuous wave propagation in optical Kerr-media. The continuous wave is x-polarized and propagates in the positive z-direction.

2d. For lossless media, solve the wave equation and give an expression for the nonlinear, intensity-dependent refractive index $n = n_0 + n_2|\mathbf{E}_\omega|^2$.

Second harmonic generation

The optical interaction

In the case of second harmonic generation (SHG), two photons at angular frequency ω combine to a photon at twice the angular frequency,

$$\hbar\omega + \hbar\omega \to \hbar(2\omega).$$

This interaction is for the second harmonic wave (at angular frequency ω)described by the second order susceptibility

$$\chi^{(2)}_{\mu\alpha\beta}(-\omega_\sigma;\omega,\omega),$$

where $\omega_\sigma = 2\omega$ is the generated second harmonic frequency of the light.

Symmetries of the medium

In this example we consider second harmonic generation in trigonal media of crystallographic point symmetry group $3m$. (Example: $LiNbO_3$)

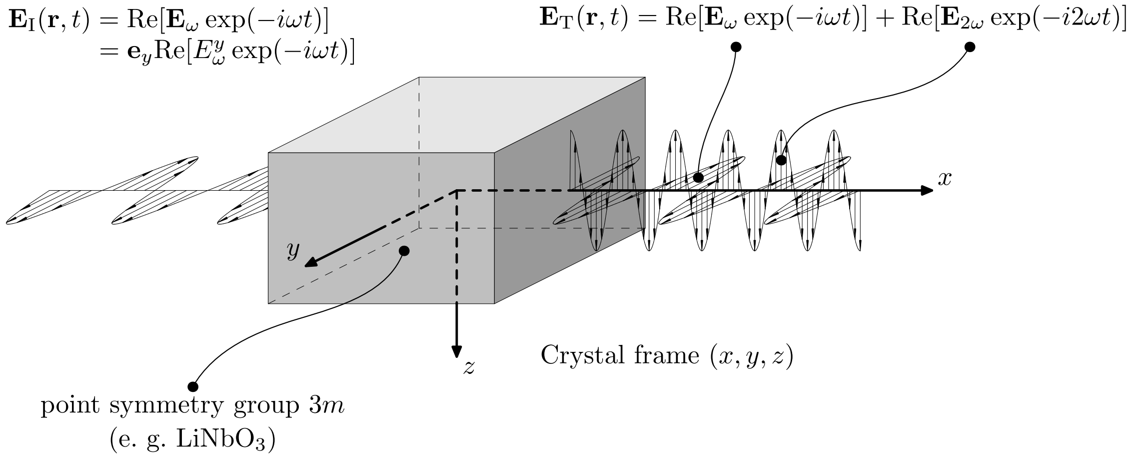

Figure 1. The setup for optical second harmonic generation in $LiNbO_3$.

For this point symmetry group, the nonzero tensor elements of the first order susceptibility are (for example according to Table A3.1 in *The Elements of Nonlinear Optics*)

$$\chi^{(1)}_{xx} = \chi^{(1)}_{yy}, \qquad \chi^{(1)}_{zz},$$

which gives the ordinary refractive indices

$$n_x(\omega) = n_y(\omega) = [1 + \chi^{(1)}_{xx}(-\omega;\omega)]^{1/2} \equiv n_\mathrm{O}(\omega)$$

for waves components polarized in the x- or y-directions, and the extraordinary refractive index

$$n_z(\omega) = [1 + \chi^{(1)}_{zz}(-\omega;\omega)]^{1/2} \equiv n_\mathrm{E}(\omega)$$

for the wave component polarized in the z-direction. Since we here are considering a negatively uniaxial crystal (see Butcher and Cotter, p. 214), these refractive indices satisfy the inequality

$$n_\mathrm{E}(\omega) \le n_\mathrm{O}(\omega).$$

The nonzero tensor elements of the second order susceptibility are (for example according to Table A3.2 in *The Elements of Nonlinear Optics*)

$$\begin{aligned} &\chi^{(2)}_{yxx} = \chi^{(2)}_{xyx} = \chi^{(2)}_{xxy} = -\chi^{(2)}_{yyy}, \qquad \chi^{(2)}_{zzz}, \\ &\chi^{(2)}_{zxx} = \chi^{(2)}_{zyy}, \qquad \chi^{(2)}_{yyz} = \chi^{(2)}_{xxz}, \qquad \chi^{(2)}_{yzy} = \chi^{(2)}_{xzx}, \end{aligned} \tag{1}$$

Additional symmetries

Intrinsic permutation symmetry for the case of second harmonic generation gives

$$\chi^{(2)}_{xxz}(-2\omega;\omega,\omega) = \chi^{(2)}_{xzx}(-2\omega;\omega,\omega) = \chi^{(2)}_{yzy}(-2\omega;\omega,\omega) = \chi^{(2)}_{yyz}(-2\omega;\omega,\omega),$$

which reduces the second order susceptibility in Eq. (1) to a set of 11 tensor elements, of which only 4 are independent. (We recall that the intrinsic permutation symmetry is always applicable, as being a consequence of the symmetrization described in lectures two and five.) Whenever Kleinman symmetry holds, the susceptibility is in addition symmetric under any permutation of the indices, which hence gives the additional relation

$$\chi^{(2)}_{zxx}(-2\omega;\omega,\omega) = \chi^{(2)}_{xzx}(-2\omega;\omega,\omega) = \chi^{(2)}_{xxz}(-2\omega;\omega,\omega),$$

i. e. reducing the second order susceptibility to a set of 11 tensor elements, of which only 3 are independent.

To summarize, the set of nonzero tensor elements describing second harmonic generation under Kleinman symmetry is

$$\begin{aligned} &\chi^{(2)}_{yxx} = \chi^{(2)}_{xyx} = \chi^{(2)}_{xxy} = -\chi^{(2)}_{yyy}, \qquad \chi^{(2)}_{zzz}, \\ &\chi^{(2)}_{zxx} = \chi^{(2)}_{zyy} = \chi^{(2)}_{yyz} = \chi^{(2)}_{xxz} = \chi^{(2)}_{yzy} = \chi^{(2)}_{xzx}. \end{aligned} \tag{2}$$

For the pump field at angular frequency ω, the relevant susceptibility describing the interaction with the second harmonic wave is[1]

$$\chi^{(2)}_{\mu\alpha\beta}(-\omega;2\omega,-\omega).$$

For an arbitrary frequency argument, this is the proper form of the susceptibility to use for the fundamental field, and this form generally differ from that of the susceptibilities for the second harmonic field. However, whenever Kleinman symmetry holds, the susceptibility for the fundamental field can be cast into the same parameters as for the second harmonic field, since

$$\begin{aligned} \chi^{(2)}_{\mu\alpha\beta}(-\omega;2\omega,-\omega) &= \{\text{Apply overall permutation symmetry}\} \\ &= \chi^{(2)}_{\alpha\mu\beta}(2\omega;-\omega,-\omega) \\ &= \{\text{Apply Kleinman symmetry}\} \\ &= \chi^{(2)}_{\mu\alpha\beta}(2\omega;-\omega,-\omega) \\ &= \{\text{Apply reality condition [B. \&C. Eq. (2.43)]}\} \\ &= [\chi^{(2)}_{\mu\alpha\beta}(-2\omega;\omega,\omega)]^* \\ &= \chi^{(2)}_{\mu\alpha\beta}(-2\omega;\omega,\omega). \end{aligned}$$

Hence the second order interaction is described by the same set of tensor elements for the fundamental as well as the second harmonic optical wave whenever Kleinman symmetry applies.

[1] Keep in mind that in the convention of Butcher and Cotter, the frequency arguments to the right of the semicolon may be writen in arbitrary order, hence we may in an equal description instead use

$$\chi^{(2)}_{xxz}(-\omega;-\omega,2\omega)$$

for the description of the second order interaction between light and matter.

The polarization density

Following the convention of Butcher and Cotter,[2] the degeneracy factor for the second harmonic signal at 2ω is

$$K(-2\omega;\omega,\omega) = 2^{l+m-n}p,$$

where

$$\begin{aligned}
p &= \{\text{the number of } \textit{distinct} \text{ permutations of } \omega,\omega\} = 1,\\
n &= \{\text{the order of the nonlinearity}\} = 2,\\
m &= \{\text{the number of angular frequencies } \omega_k \text{ that are zero}\} = 0,\\
l &= \left\{\begin{array}{ll} 1, & \text{if } 2\omega \neq 0,\\ 0, & \text{otherwise.}\end{array}\right\} = 1,
\end{aligned}$$

i. e.

$$K(-2\omega;\omega,\omega) = 2^{1+0-2}\times 1 = 1/2.$$

For the fundamental optical field at ω, one might be mislead to assume that since the second order interaction for this field is described by an identical set of tensor elements as for the second harmonic wave, the degeneracy factor must also be identical to the previously derived one. This is, however, *a very wrong assumption*, and one can easily verify that the proper degeneracy factor for the fundamental field instead is given as

$$K(-\omega;2\omega,-\omega) = 2^{l+m-n}p,$$

where

$$\begin{aligned}
p &= \{\text{the number of } \textit{distinct} \text{ permutations of } 2\omega,-\omega\} = 2,\\
n &= \{\text{the order of the nonlinearity}\} = 2,\\
m &= \{\text{the number of angular frequencies } \omega_k \text{ that are zero}\} = 0,\\
l &= \left\{\begin{array}{ll} 1, & \text{if } \omega \neq 0,\\ 0, & \text{otherwise.}\end{array}\right\} = 1,
\end{aligned}$$

i. e.

$$K(-\omega;2\omega,-\omega) = 2^{1+0-2}\times 2 = 1.$$

The general second harmonic polarization density of the medium is hence given as

$$\begin{aligned}
[\mathbf{P}^{(\mathrm{NL})}_{2\omega}]_z = [\mathbf{P}^{(2)}_{2\omega}]_z &= \varepsilon_0 \underbrace{K(-2\omega;\omega,\omega)\chi^{(2)}_{z\alpha\beta}(-2\omega;\omega,\omega)}_{=\frac{1}{2}\chi^{(2)}_{z\alpha\beta}(-2\omega;\omega,\omega)} E^{\alpha}_{\omega}E^{\beta}_{\omega}\\
&= (\varepsilon_0/2)[\chi^{(2)}_{zxx}E^x_\omega E^x_\omega + \chi^{(2)}_{zyy}E^y_\omega E^y_\omega + \chi^{(2)}_{zzz}E^z_\omega E^z_\omega]\\
&= (\varepsilon_0/2)[\chi^{(2)}_{zxx}(E^x_\omega E^x_\omega + E^y_\omega E^y_\omega) + \chi^{(2)}_{zzz}E^z_\omega E^z_\omega],\\
[\mathbf{P}^{(\mathrm{NL})}_{2\omega}]_y &= (\varepsilon_0/2)[\chi^{(2)}_{yxx}E^x_\omega E^x_\omega + \chi^{(2)}_{yyy}E^y_\omega E^y_\omega + \chi^{(2)}_{yyz}E^y_\omega E^z_\omega + \chi^{(2)}_{yzy}E^z_\omega E^y_\omega]\\
&= (\varepsilon_0/2)[\chi^{(2)}_{yxx}(E^x_\omega E^x_\omega - E^y_\omega E^y_\omega) + \chi^{(2)}_{zxx}(E^y_\omega E^z_\omega + E^z_\omega E^y_\omega)],\\
[\mathbf{P}^{(\mathrm{NL})}_{2\omega}]_x &= (\varepsilon_0/2)[\chi^{(2)}_{xxy}E^x_\omega E^y_\omega + \chi^{(2)}_{xyx}E^y_\omega E^x_\omega + \chi^{(2)}_{xxz}E^x_\omega E^z_\omega + \chi^{(2)}_{xzx}E^z_\omega E^x_\omega]\\
&= (\varepsilon_0/2)[\chi^{(2)}_{yxx}(E^x_\omega E^y_\omega + E^y_\omega E^x_\omega) + \chi^{(2)}_{zxx}(E^x_\omega E^z_\omega + E^z_\omega E^x_\omega)],
\end{aligned}$$

[2] See course material on the Butcher and Cotter convention handed out during the third lecture. Notice that for the first order polarization density, one at optical frequencies *always* has the trivial degeneracy factor

$$K(-2\omega;\omega) = 2^{l+m-n}p = 2^{1+0-1}\times 1 = 1.$$

while the general polarization density at the angular frequency of the pump field becomes[3]

$$
\begin{aligned}
[\mathbf{P}^{(\mathrm{NL})}_{\omega}]_z = [\mathbf{P}^{(2)}_{\omega}]_z &= \varepsilon_0 \underbrace{K(-\omega;2\omega,-\omega)\chi^{(2)}_{z\alpha\beta}(-\omega;2\omega,-\omega)}_{=\chi^{(2)}_{z\alpha\beta}(-2\omega;\omega,\omega)} E^{\alpha}_{2\omega}E^{\beta}_{-\omega}\\
&= \varepsilon_0[\chi^{(2)}_{zxx}E^x_{2\omega}E^{x*}_{\omega} + \chi^{(2)}_{zyy}E^y_{2\omega}E^{y*}_{\omega} + \chi^{(2)}_{zzz}E^z_{2\omega}E^{z*}_{\omega}]\\
&= \varepsilon_0[\chi^{(2)}_{zxx}(E^x_{2\omega}E^{x*}_{\omega} + E^y_{2\omega}E^{y*}_{\omega}) + \chi^{(2)}_{zzz}E^z_{2\omega}E^{z*}_{\omega}],\\
[\mathbf{P}^{(\mathrm{NL})}_{\omega}]_y &= \varepsilon_0[\chi^{(2)}_{yxx}E^x_{2\omega}E^{x*}_{\omega} + \chi^{(2)}_{yyy}E^y_{2\omega}E^{y*}_{\omega} + \chi^{(2)}_{yyz}E^y_{2\omega}E^{z*}_{\omega} + \chi^{(2)}_{yzy}E^z_{2\omega}E^{y*}_{\omega}]\\
&= \varepsilon_0[\chi^{(2)}_{yxx}(E^x_{2\omega}E^{x*}_{\omega} - E^y_{2\omega}E^{y*}_{\omega}) + \chi^{(2)}_{zxx}(E^y_{2\omega}E^{z*}_{\omega} + E^z_{2\omega}E^{y*}_{\omega})],\\
[\mathbf{P}^{(\mathrm{NL})}_{\omega}]_x &= \varepsilon_0[\chi^{(2)}_{xxy}E^x_{2\omega}E^{y*}_{\omega} + \chi^{(2)}_{xyx}E^y_{2\omega}E^{x*}_{\omega} + \chi^{(2)}_{xxz}E^x_{2\omega}E^{z*}_{\omega} + \chi^{(2)}_{xzx}E^z_{2\omega}E^{x*}_{\omega}]\\
&= \varepsilon_0[\chi^{(2)}_{yxx}(E^x_{2\omega}E^{y*}_{\omega} + E^y_{2\omega}E^{x*}_{\omega}) + \chi^{(2)}_{zxx}(E^x_{2\omega}E^{z*}_{\omega} + E^z_{2\omega}E^{x*}_{\omega})].
\end{aligned}
$$

For a pump wave polarized in the yz-plane of the crystal frame, the polarization density of the medium hence becomes

$$
\begin{aligned}
[\mathbf{P}^{(\mathrm{NL})}_{2\omega}]_z &= (\varepsilon_0/2)[\chi^{(2)}_{zxx}E^y_{\omega}E^y_{\omega} + \chi^{(2)}_{zzz}E^z_{\omega}E^z_{\omega}],\\
[\mathbf{P}^{(\mathrm{NL})}_{2\omega}]_y &= (\varepsilon_0/2)[-\chi^{(2)}_{yxx}E^y_{\omega}E^y_{\omega} + \chi^{(2)}_{zxx}(E^y_{\omega}E^z_{\omega} + E^z_{\omega}E^y_{\omega})],\\
[\mathbf{P}^{(\mathrm{NL})}_{2\omega}]_x &= 0,
\end{aligned}
$$

and

$$
\begin{aligned}
[\mathbf{P}^{(\mathrm{NL})}_{\omega}]_z &= \varepsilon_0[\chi^{(2)}_{zxx}E^y_{2\omega}E^{y*}_{\omega} + \chi^{(2)}_{zzz}E^z_{2\omega}E^{z*}_{\omega}],\\
[\mathbf{P}^{(\mathrm{NL})}_{\omega}]_y &= \varepsilon_0[-\chi^{(2)}_{yxx}E^y_{2\omega}E^{y*}_{\omega} + \chi^{(2)}_{zxx}(E^y_{2\omega}E^{z*}_{\omega} + E^z_{2\omega}E^{y*}_{\omega})],\\
[\mathbf{P}^{(\mathrm{NL})}_{\omega}]_x &= 0.
\end{aligned}
$$

The wave equation

Strictly speaking, the previously formulated polarization density gives a coupled system between the polarization states of both the fundamental and second harmonic waves, since both the y- and z-components of the polarization densities at ω and 2ω contain components of all other field components. However, for simplicity we will here restrict the continued analysis to the case of a y-polarized input pump wave, which through the $\chi^{(2)}_{zyy} = \chi^{(2)}_{zxx}$ elements give rise to a z-polarized second harmonic frequency component at 2ω.

The electric fields of the fundamental and second harmonic optical waves are for the forward propagating configuration expressed in their *spatial envelopes* $\mathbf{A}_{\omega}$ and $\mathbf{A}_{2\omega}$ as

$$
\begin{aligned}
\mathbf{E}_{\omega}(x) &= \mathbf{e}_y A^y_{\omega}(x)\exp(ik_{\omega_y}x), \qquad k_{\omega_y} \equiv \omega n_{\omega_y}/c \equiv \omega n_{\mathrm{O}}(\omega)/c\\
\mathbf{E}_{2\omega}(x) &= \mathbf{e}_z A^z_{2\omega}(x)\exp(ik_{2\omega_z}x), \qquad k_{2\omega_z} \equiv 2\omega n_{2\omega_z}/c \equiv 2\omega n_{\mathrm{E}}(2\omega)/c
\end{aligned}
$$

Using the above separation of the natural, spatial oscillation of the light, in the infinite plane wave approximation and by using the slowly varying envelope approximation, the wave equation for the envelope of the second harmonic optical field becomes (see Eq. (6) in the notes from lecture eight)

$$
\begin{aligned}
\frac{\partial A^z_{2\omega}}{\partial x} &= i\frac{\mu_0(2\omega)^2}{2k_{2\omega_z}}[\mathbf{P}^{(\mathrm{NL})}_{2\omega}]_z\exp(-ik_{2\omega_z}x)\\
&= i\frac{\mu_0(2\omega)^2}{2(2\omega n_{2\omega_z}/c)}\underbrace{\frac{\varepsilon_0}{2}\chi^{(2)}_{zxx}A^{y\,2}_{\omega}\exp(2ik_{\omega_y}x)}_{=[\mathbf{P}^{(\mathrm{NL})}_{2\omega}]_z}\exp(-ik_{2\omega_z}x)\\
&= i\frac{\omega\chi^{(2)}_{zxx}}{2n_{2\omega_z}c}A^{y\,2}_{\omega}\exp[i(2k_{\omega_y} - k_{2\omega_z})x],
\end{aligned}
$$

[3] Keep in mind that a negative frequency argument to the right of the semicolon in the susceptibility is to be associated with the complex conjugate of the respective electric field; see Butcher and Cotter, section 2.3.2.

while for the fundamental wave,

$$\begin{aligned}\frac{\partial A_\omega^y}{\partial x} &= i\frac{\mu_0\omega^2}{2k_{\omega_y}}[\mathbf{P}_\omega^{(\mathrm{NL})}]_y \exp(-ik_{\omega_y}x)\\ &= i\frac{\mu_0\omega^2}{2(\omega n_{\omega_y}/c)}\underbrace{\varepsilon_0\chi_{zxx}^{(2)}A_{2\omega}^z\exp(ik_{2\omega_z}x)A_\omega^{y*}\exp(-ik_{\omega_y}x)}_{=[\mathbf{P}_\omega^{(\mathrm{NL})}]_y}\exp(-ik_{\omega_y}x)\\ &= i\frac{\omega\chi_{zxx}^{(2)}}{2n_{\omega_y}c}A_{2\omega}^z A_\omega^{y*}\exp[-i(2k_{\omega_y}-k_{2\omega_z})x].\end{aligned}$$

These equations can hence be summarized by the coupled system

$$\frac{\partial A_{2\omega}^z}{\partial x} = i\frac{\omega\chi_{zxx}^{(2)}}{2n_{2\omega_z}c}A_\omega^{y\,2}\exp(i\Delta kx), \tag{3a}$$

$$\frac{\partial A_\omega^y}{\partial x} = i\frac{\omega\chi_{zxx}^{(2)}}{2n_{\omega_y}c}A_{2\omega}^z A_\omega^{y*}\exp(-i\Delta kx). \tag{3b}$$

where

$$\begin{aligned}\Delta k &= 2k_{\omega_y} - k_{2\omega_z}\\ &= 2\omega n_{\omega_y}/c - 2\omega n_{2\omega_z}/c\\ &= (2\omega/c)(n_{\omega_y} - \omega n_{2\omega_z})\end{aligned}$$

is the so-called phase mismatch between the pump and second harmonic wave.

Boundary conditions

Here the boundary conditions are simply that no second harmonic signal is present at the input,

$$A_{2\omega}^z(0) = 0,$$

together with a known input field at the fundamental frequency,

$$A_\omega^y(0) = \{\text{known}\}.$$

Solving the wave equation

Considering a nonzero Δk, the conversion efficiency is regularly quite small, and one may approximately take the spatial distribution of the pump wave to be constant, $A_\omega^y(x) \approx A_\omega^y(0)$. Using this approximation[4], and by applying the initial condition $A_{2\omega}^z(0) = 0$ of the second harmonic signal, one finds

$$\begin{aligned}A_{2\omega}^z(L) &= \int_0^L \frac{\partial A_{2\omega}^z(z)}{\partial x}\,dx\\ &= \int_0^L i\frac{\omega\chi_{zxx}^{(2)}}{2n_{2\omega_z}c}A_\omega^{y\,2}(0)\exp(i\Delta kx)\,dx\\ &= \frac{\omega\chi_{zxx}^{(2)}}{2n_{2\omega_z}c}A_\omega^{y\,2}(0)\frac{1}{\Delta k}[\exp(i\Delta kL)-1]\\ &= \big\{\text{Use } [\exp(i\Delta kL)-1]/\Delta k = iL\exp(i\Delta kL/2)\operatorname{sinc}(\Delta kL/2)\big\}\\ &= i\frac{\omega\chi_{zxx}^{(2)}L}{2n_{2\omega_z}c}A_\omega^{y\,2}(0)\exp(i\Delta kL/2)\operatorname{sinc}(\Delta kL/2).\end{aligned}$$

[4] For an outline of the method of solving the coupled system (1) exactly in terms of Jacobian elliptic functions (thus allowing for a depleted pump as well), see J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, Phys. Rev. **127**, 1918–1939, (1962).

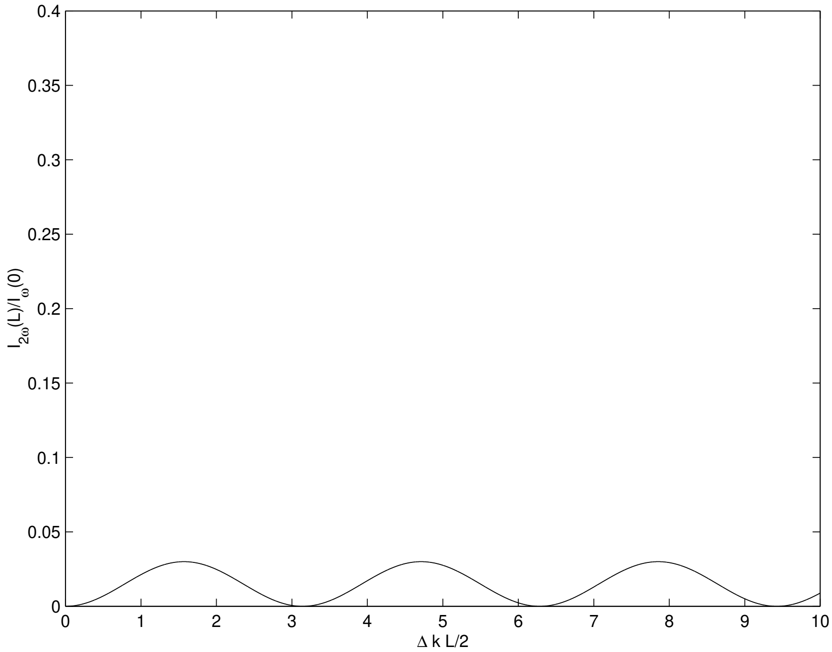

Figure 2. Conversion efficiency $I_{2\omega}(L)/I_\omega(0)$ as function of normalized crystal length $\Delta kL/2$. The conversion efficiency is in the phase mismatched case ($\Delta k \neq 0$) a periodic function, with period $2L_{\mathrm{c}}$, with $L_{\mathrm{c}} = \pi/\Delta k$ being the *coherence length*.

In terms if the light intensities of the waves, one after a propagation distance $x = L$ hence has the second harmonic signal with intensity $I_{2\omega}(L)$ expressed in terms of the input intensity I_ω as

$$
\begin{aligned}
I_{2\omega}(L) &= \frac{1}{2}\varepsilon_0 c n_{2\omega_z} |A^z_{2\omega}(L)|^2 \\
&= \frac{1}{2}\varepsilon_0 c n_{2\omega_z} \Big| i\frac{\omega\chi^{(2)}_{zxx}L}{2n_{2\omega_z}c} A^y_\omega{}^2(0)\exp(i\Delta kL/2)\,\mathrm{sinc}(\Delta kL/2)\Big|^2 \\
&= \varepsilon_0 \frac{\omega^2 L^2}{8n_{2\omega_z}c} |\chi^{(2)}_{zxx}|^2 |A^y_\omega(0)|^4 \,\mathrm{sinc}^2(\Delta kL/2) \\
&= \left\{ \text{Use } |A^y_\omega(0)|^2 = \frac{2I_\omega(0)}{\varepsilon_0 c n_{\omega_y}} \right\} \\
&= \varepsilon_0 \frac{\omega^2 L^2}{8n_{2\omega_z}c} |\chi^{(2)}_{zxx}|^2 \frac{4I^2_\omega(0)}{\varepsilon_0^2 c^2 n^2_{\omega_y}} \,\mathrm{sinc}^2(\Delta kL/2) \\
&= \frac{\omega^2 L^2}{2\varepsilon_0 c^3} \frac{|\chi^{(2)}_{zxx}(-2\omega;\omega,\omega)|^2}{n_{2\omega_z} n^2_{\omega_y}} I^2_\omega(0)\,\mathrm{sinc}^2(\Delta kL/2),
\end{aligned}
$$

i. e. with the conversion efficiency

$$
\frac{I_{2\omega}(L)}{I_\omega(0)} = \frac{\omega^2 L^2}{2\varepsilon_0 c^3} \frac{|\chi^{(2)}_{zxx}(-2\omega;\omega,\omega)|^2}{n_{2\omega_z} n^2_{\omega_y}} I_\omega(0)\,\mathrm{sinc}^2(\Delta kL/2).
$$

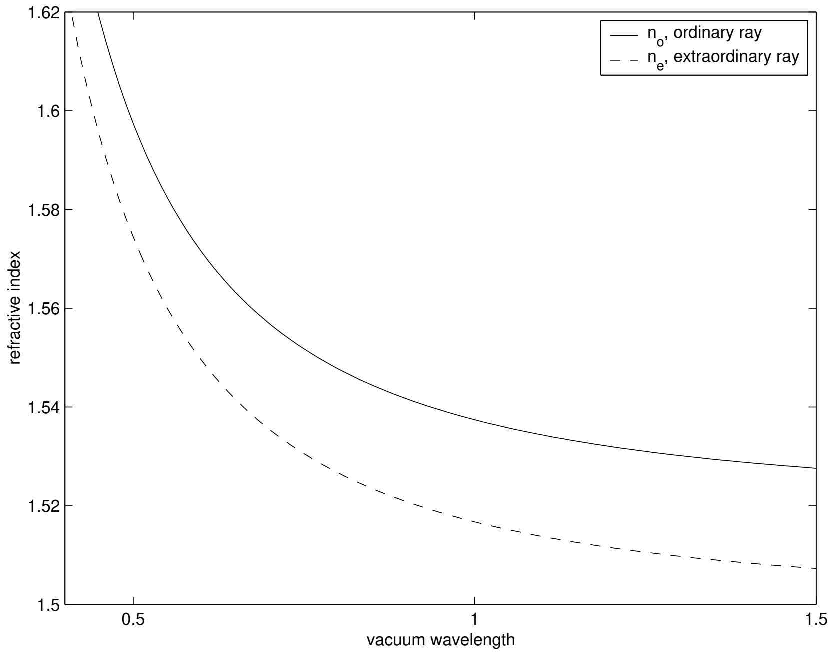

Figure 3. Ordinary and extraordinary refractive indices of a negative uniaxial crystal as function of vacuum wavelength of the light, in the case of normal dispersion. Phase matching between the pump and second harmonic wave is obtained whenever $n_{\omega_y} \equiv n_{\mathrm{O}}(\omega) = n_{\mathrm{E}}(2\omega) \equiv n_{2\omega_z}$.

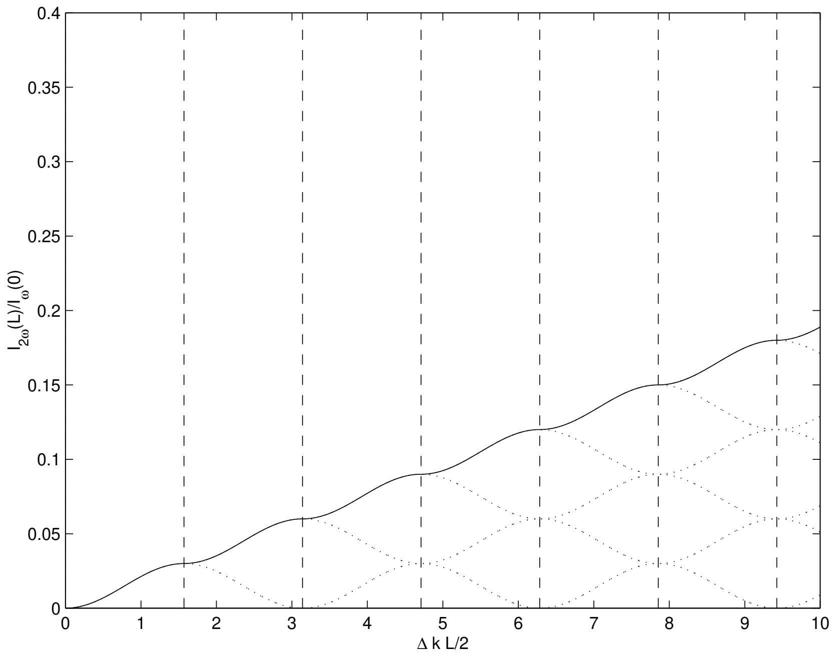

Figure 4. Conversion efficiency $I_{2\omega}(L)/I_{\omega}(0)$ as function of normalized crystal length $\Delta kL/2$ when the material properties are periodically reversed, with a half-period of L_{c}.

Optical Kerr-effect - Field corrected refractive index

As a start, we assume a monochromatic optical wave (containing forward and/or backward propagating components) polarized in the xy-plane,

$$\mathbf{E}(z,t) = \mathrm{Re}[\mathbf{E}_\omega(z)\exp(-i\omega t)] \in \mathbb{R}^3,$$

with all spatial variation of the field contained in

$$\mathbf{E}_\omega(z) = \mathbf{e}_x E_\omega^x(z) + \mathbf{e}_y E_\omega^y(z) \in \mathbb{C}^3.$$

The optical interaction

Optical Kerr-effect is in isotropic media described by the third order susceptibility[5]

$$\chi^{(3)}_{\mu\alpha\beta\gamma}(-\omega;\omega,\omega,-\omega).$$

Symmetries of the medium

The general set of nonzero components of $\chi^{(3)}_{\mu\alpha\beta\gamma}$ for isotropic media are from Appendix A3.3 of Butcher and Cotters book given as

$$\begin{aligned}
&\chi^{(3)}_{xxxx} = \chi^{(3)}_{yyyy} = \chi^{(3)}_{zzzz},\\
&\chi^{(3)}_{yyzz} = \chi^{(3)}_{zzyy} = \chi^{(3)}_{zzxx} = \chi^{(3)}_{xxzz} = \chi^{(3)}_{xxyy} = \chi^{(3)}_{yyxx}\\
&\chi^{(3)}_{yzyz} = \chi^{(3)}_{zyzy} = \chi^{(3)}_{zxzx} = \chi^{(3)}_{xzxz} = \chi^{(3)}_{xyxy} = \chi^{(3)}_{yxyx}\\
&\chi^{(3)}_{yzzy} = \chi^{(3)}_{zyyz} = \chi^{(3)}_{zxxz} = \chi^{(3)}_{xzzx} = \chi^{(3)}_{xyyx} = \chi^{(3)}_{yxxy}
\end{aligned}\tag{4}$$

with

$$\chi^{(3)}_{xxxx} = \chi^{(3)}_{xxyy} + \chi^{(3)}_{xyxy} + \chi^{(3)}_{xyyx},$$

i. e. a general set of 21 elements, of which only 3 are independent.

Additional symmetries

By applying the intrinsic permutation symmetry in the middle indices for optical Kerr-effect, one generally has

$$\chi^{(3)}_{\mu\alpha\beta\gamma}(-\omega;\omega,\omega,-\omega) = \chi^{(3)}_{\mu\beta\alpha\gamma}(-\omega;\omega,\omega,-\omega),$$

which hence slightly reduce the set (4) to still 21 nonzero elements, but of which now only two are independent. For a beam polarized in the xy-plane, the elements of interest are only those which only contain x or y in the indices, i. e. the subset

$$\chi^{(3)}_{xxxx} = \chi^{(3)}_{yyyy}, \quad \chi^{(3)}_{xxyy} = \chi^{(3)}_{yyxx} = \left\{ \begin{array}{c} \text{intr. perm. symm.} \\ (\alpha,\omega) \rightleftharpoons (\beta,\omega) \end{array} \right\} = \chi^{(3)}_{xyxy} = \chi^{(3)}_{yxyx}, \quad \chi^{(3)}_{xyyx} = \chi^{(3)}_{yxxy},$$

with

$$\chi^{(3)}_{xxxx} = \chi^{(3)}_{xxyy} + \chi^{(3)}_{xyxy} + \chi^{(3)}_{xyyx},$$

i. e. a set of eight elements, of which only two are independent.

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[5] Again, keep in mind that in the convention of Butcher and Cotter, the frequency arguments to the right of the semicolon may be writen in arbitrary order, hence we may in an equal description instead use

$$\chi^{(3)}_{\mu\alpha\beta\gamma}(-\omega;\omega,-\omega,\omega)$$

or

$$\chi^{(3)}_{\mu\alpha\beta\gamma}(-\omega;-\omega,\omega,\omega)$$

for this description of the third order interaction between light and matter.

The polarization density

The degeneracy factor $K(-\omega;\omega,\omega,-\omega)$ is calculated as

$$K(-\omega;\omega,\omega,-\omega)=2^{l+m-n}p=2^{1+0-3}3=3/4.$$

From the reduced set of nonzero susceptibilities for the beam polarized in the xy-plane, and by using the calculated value of the degeneracy factor in the convention of Butcher and Cotter, we hence have the third order electric polarization density at $\omega_\sigma=\omega$ given as $\mathbf{P}^{(n)}(\mathbf{r},t)=\mathrm{Re}[\mathbf{P}_\omega^{(n)}\exp(-i\omega t)]$, with

$$\begin{aligned}
\mathbf{P}_\omega^{(3)} &= \sum_\mu \mathbf{e}_\mu (P_\omega^{(3)})_\mu \\
&= \{\text{Using the convention of Butcher and Cotter}\} \\
&= \sum_\mu \mathbf{e}_\mu \Big[\varepsilon_0 \frac{3}{4}\sum_\alpha\sum_\beta\sum_\gamma \chi^{(3)}_{\mu\alpha\beta\gamma}(-\omega;\omega,\omega,-\omega)(E_\omega)_\alpha(E_\omega)_\beta(E_{-\omega})_\gamma\Big] \\
&= \{\text{Evaluate the sums over } (x,y,z) \text{ for field polarized in the } xy \text{ plane}\} \\
&= \varepsilon_0\frac{3}{4}\{\mathbf{e}_x[\chi^{(3)}_{xxxx}E^x_\omega E^x_\omega E^x_{-\omega}+\chi^{(3)}_{xyyx}E^y_\omega E^y_\omega E^x_{-\omega}+\chi^{(3)}_{xyxy}E^y_\omega E^x_\omega E^y_{-\omega}+\chi^{(3)}_{xxyy}E^x_\omega E^y_\omega E^y_{-\omega}] \\
&\qquad +\mathbf{e}_y[\chi^{(3)}_{yyyy}E^y_\omega E^y_\omega E^y_{-\omega}+\chi^{(3)}_{yxxy}E^x_\omega E^x_\omega E^y_{-\omega}+\chi^{(3)}_{yxyx}E^x_\omega E^y_\omega E^x_{-\omega}+\chi^{(3)}_{yyxx}E^y_\omega E^x_\omega E^x_{-\omega}]\} \\
&= \{\text{Make use of } \mathbf{E}_{-\omega}=\mathbf{E}^*_\omega \text{ and relations } \chi^{(3)}_{xxyy}=\chi^{(3)}_{yyxx},\ \text{etc.}\} \\
&= \varepsilon_0\frac{3}{4}\{\mathbf{e}_x[\chi^{(3)}_{xxxx}E^x_\omega|E^x_\omega|^2+\chi^{(3)}_{xyyx}E^{y\,2}_\omega E^{x*}_\omega+\chi^{(3)}_{xyxy}|E^y_\omega|^2E^x_\omega+\chi^{(3)}_{xxyy}E^x_\omega|E^y_\omega|^2] \\
&\qquad +\mathbf{e}_y[\chi^{(3)}_{xxxx}E^y_\omega|E^y_\omega|^2+\chi^{(3)}_{xyyx}E^{x\,2}_\omega E^{y*}_\omega+\chi^{(3)}_{xyxy}|E^x_\omega|^2E^y_\omega+\chi^{(3)}_{xxyy}E^y_\omega|E^x_\omega|^2]\} \\
&= \{\text{Make use of the intrinsic permutation symmetry}\} \\
&= \varepsilon_0\frac{3}{4}\{\mathbf{e}_x[(\chi^{(3)}_{xxxx}|E^x_\omega|^2+2\chi^{(3)}_{xxyy}|E^y_\omega|^2)E^x_\omega+(\chi^{(3)}_{xxxx}-2\chi^{(3)}_{xxyy})E^{y\,2}_\omega E^{x*}_\omega \\
&\qquad \mathbf{e}_y[(\chi^{(3)}_{xxxx}|E^y_\omega|^2+2\chi^{(3)}_{xxyy}|E^x_\omega|^2)E^y_\omega+(\chi^{(3)}_{xxxx}-2\chi^{(3)}_{xxyy})E^{x\,2}_\omega E^{y*}_\omega.
\end{aligned}$$

For the optical field being linearly polarized, say in the x-direction, the expression for the polarization density is significantly simplified, to yield

$$\mathbf{P}_\omega^{(3)}=\varepsilon_0(3/4)\mathbf{e}_x\chi^{(3)}_{xxxx}|E^x_\omega|^2E^x_\omega,$$

i. e. taking a form that can be interpreted as an intensity-dependent ($\sim|E^x_\omega|^2$) contribution to the refractive index (cf. Butcher and Cotter §6.3.1).

The wave equation – Time independent case

In this example, we consider continuous wave propagation[6] in optical Kerr-media, using light polarized in the x-direction and propagating along the positive direction of the z-axis,

$$\mathbf{E}(\mathbf{r},t)=\mathrm{Re}[\mathbf{E}_\omega(z)\exp(-i\omega t)],\qquad \mathbf{E}_\omega(z)=\mathbf{A}_\omega(z)\exp(ikz)=\mathbf{e}_xA^x_\omega(z)\exp(ikz),$$

where, as previously, $k=\omega n_0/c$. From material handed out during the third lecture (notes on the Butcher and Cotter convention), the nonlinear polarization density for x-polarized light is given as $\mathbf{P}_\omega^{(\mathrm{NL})}=\mathbf{P}_\omega^{(3)}$, with

$$\begin{aligned}
\mathbf{P}_\omega^{(3)} &= \varepsilon_0(3/4)\mathbf{e}_x\chi^{(3)}_{xxxx}(-\omega;\omega,\omega,-\omega)|E^x_\omega|^2E^x_\omega \\
&= \varepsilon_0(3/4)\chi^{(3)}_{xxxx}|\mathbf{E}_\omega|^2\mathbf{E}_\omega \\
&= \varepsilon_0(3/4)\chi^{(3)}_{xxxx}|\mathbf{A}_\omega|^2\mathbf{A}_\omega\exp(ikz),
\end{aligned}$$

[6] That is to say, a time independent problem with the temporal envelope of the electrical field being constant in time.

and the time independent wave equation for the field envelope $\mathbf{A}_\omega$, using Eq. (6), becomes

$$\begin{aligned}
\frac{\partial}{\partial z}\mathbf{A}_\omega &= i\frac{\mu_0\omega^2}{2k}\underbrace{\varepsilon_0(3/4)\chi^{(3)}_{xxxx}|\mathbf{A}_\omega|^2\mathbf{A}_\omega\exp(ikz)}_{=\mathbf{P}^{(\mathrm{NL})}_\omega(z)}\exp(-ikz)\\
&= i\frac{3\omega^2}{8c^2k}\chi^{(3)}_{xxxx}|\mathbf{A}_\omega|^2\mathbf{A}_\omega\\
&= \{\text{since } k=\omega n_0(\omega)/c\}\\
&= i\frac{3\omega}{8cn_0}\chi^{(3)}_{xxxx}|\mathbf{A}_\omega|^2\mathbf{A}_\omega,
\end{aligned}$$

or, equivalently, in its scalar form

$$\frac{\partial}{\partial z}A^x_\omega = i\frac{3\omega}{8cn_0}\chi^{(3)}_{xxxx}|A^x_\omega|^2A^x_\omega.$$

Boundary conditions – Time independent case

For this special case of unidirectional wave propagation, the boundary condition is simply a known optical field at the input,

$$A^x_\omega(0) = \{\text{known}\}.$$

Solving the wave equation – Time independent case

If the medium of interest now is analyzed at an angular frequency far from any resonance, we may look for solutions to this equation with $|\mathbf{A}_\omega(z)|$ being constant (for a lossless medium). For such a case it is straightforward to integrate the final wave equation to yield the general solution

$$\mathbf{A}_\omega(z) = \mathbf{A}_\omega(0)\exp[i\frac{3\omega}{8cn_0}\chi^{(3)}_{xxxx}|\mathbf{A}_\omega(0)|^2z],$$

or, again equivalently, in the scalar form

$$A^x_\omega(z) = A^x_\omega(0)\exp[i\frac{3\omega}{8cn_0}\chi^{(3)}_{xxxx}|A^x_\omega(0)|^2z],$$

which hence gives the solution for the real-valued electric field $\mathbf{E}(\mathbf{r},t)$ as

$$\begin{aligned}
\mathbf{E}(\mathbf{r},t) &= \mathrm{Re}[\mathbf{E}_\omega(z)\exp(-i\omega t)]\\
&= \mathrm{Re}\{\mathbf{A}_\omega(z)\exp[i(kz-\omega t)]\}\\
&= \mathrm{Re}\{\mathbf{A}_\omega(0)\exp[i(\underbrace{\frac{\omega n_0}{c}z+\frac{3\omega}{8cn_0}\chi^{(3)}_{xxxx}|A^x_\omega(0)|^2z}_{\equiv k_{\mathrm{eff}}z}-\omega t)]\}.
\end{aligned}$$

From this solution, one immediately finds that the wave propagates with an effective propagation constant

$$k_{\mathrm{eff}} = \frac{\omega}{c}[n_0+\frac{3}{8n_0}\chi^{(3)}_{xxxx}|A^x_\omega(0)|^2],$$

that is to say, experiencing the intensity dependent refractive index

$$\begin{aligned}
n_{\mathrm{eff}} &= n_0+\frac{3}{8n_0}\chi^{(3)}_{xxxx}|A^x_\omega(0)|^2\\
&= n_0+n_2|A^x_\omega(0)|^2,
\end{aligned}$$

with

$$n_2 = \frac{3}{8n_0}\chi^{(3)}_{xxxx}.$$