Nonlinear Electromagnetism
Tutorial n°3
Second harmonic intensity optimization in a uniaxial crystal

A plane wave, at frequency ω is incident normally on the input face of a uniaxial crystal, with a second order nonlinear susceptibility and we are interested in optimizing the second harmonic generation at 2ω in this crystal. The wavevector of the incident wave is defined by its Euler angles θ and ϕ in the $(Oxyz)$ coordinate system.

The incident field is decomposed into the sum of ordinary and extraordinary vibration modes.

$$\boldsymbol{E}(\omega) = \boldsymbol{E}_o(\omega) + \boldsymbol{E}_\theta(\omega).$$

with

$$\begin{aligned} \boldsymbol{E}_o(\omega) &= A_o(\omega)\boldsymbol{e}_o \exp i(\boldsymbol{k}_o(\omega).\boldsymbol{s}), \\ \boldsymbol{E}_\theta(\omega) &= A_\theta(\omega)\boldsymbol{e}_\theta \exp i(\boldsymbol{k}_\theta(\omega).\boldsymbol{s}). \end{aligned}$$

Only these two eigen modes can propagate in the uniaxial crystal without any polarization change. Although these two components propagate at different speed, ordinary and extraordinary indices are close enough to assume that $\boldsymbol{D}$ is parallel to$\boldsymbol{E}$,

In the same way, the wave at 2ω, propagating in the same direction can be written:

$$\boldsymbol{E}(2\omega) = \boldsymbol{E}_o(2\omega) + \boldsymbol{E}_\theta(2\omega)$$

with

$$\begin{aligned} \boldsymbol{E}_o(2\omega) &= A_o(2\omega)\boldsymbol{e}_o \exp i(\boldsymbol{k}_o(2\omega).\boldsymbol{s}), \\ \boldsymbol{E}_\theta(2\omega) &= A_\theta(2\omega)\boldsymbol{e}_\theta \exp i(\boldsymbol{k}_\theta(2\omega).\boldsymbol{s}). \end{aligned}$$

The propagation modes $\boldsymbol{E}_o(2\omega)$ et $\boldsymbol{E}_\theta(2\omega)$ are orthogonal and therefore do not interfere and the nonlinear wave equation can be projected on directions $\boldsymbol{e}_o$ et $\boldsymbol{e}_\theta$

$$\begin{cases} \frac{\partial A_o(2\omega)}{\partial s} = \frac{i(2\omega)}{2nc\epsilon_o}\boldsymbol{e}_o.\boldsymbol{P}_{NL}(2\omega)\exp(-ik_o(2\omega)s) \\ \frac{\partial A_\theta(2\omega)}{\partial s} = \frac{i(2\omega)}{2nc\epsilon_o}\boldsymbol{e}_\theta.\boldsymbol{P}_{NL}(2\omega)\exp(-ik_\theta(2\omega)s) \end{cases}$$

The optimization of the second harmonic generation process requires

- phase matching between the nonlinear polarisation and the propagating wave at 2ω
- optimize the nonlinear polarization

1. **Eigenmodes $\boldsymbol{e}_o$ et $\boldsymbol{e}_\theta$ in a uniaxial crystal**

   (a) Using Fresnel equation, show that the index experienced by the wave propagating in direction $\boldsymbol{k}(\theta,\phi)$ is given by the following equation:

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

   (b) Show that the ordinary and extraordinary vibration directions are respectively:

   $$\boldsymbol{e}_o \left| \begin{matrix} \sin\phi \\ -\cos\phi \\ 0 \end{matrix} \right. , \quad \boldsymbol{e}_\theta \left| \begin{matrix} -\cos\theta\cos\phi \\ -\cos\theta\sin\phi \\ \sin\theta \end{matrix} \right. .$$

2. **Incident field amplitude** The incident field is assumed to have an arbitrary elliptical polarization state with respect to the axes $\boldsymbol{e}_X$ and $\boldsymbol{e}_Y$. At $s = 0$ it can be expressed in the following way:

$$\boldsymbol{E}(\omega) = \frac{\sqrt{2}}{2} A \left(\cos(\varphi/2) \boldsymbol{e}_X - i \sin(\varphi/2) \boldsymbol{e}_Y \right).$$

$\boldsymbol{E}(\omega)$ is decomposed into eigen vibration modes $\boldsymbol{E}(\omega) = A_o \boldsymbol{e}_o + A_\theta \boldsymbol{e}_\theta$. Give the expressions of the amplitudes A_o and A_θ

3. **Nonlinear wave equations**

   (a) Give the expression of the nonlinear polarization $\boldsymbol{P}_{NL}(2\omega)$.

   (b) Write the nonlinear wave equations, taking into account the result of the previous question.

4. **Phase matching** Considering both cases of uniaxial positive ($n_e > n_o$) and negative ($n_o > n_e$) crystals, write the phase matching conditions for ordinary and extraordinary propagation modes at frequency 2ω. What can be said ?

Definition: *When the phase matching condition is satisfied through a relation like* $n_i(\omega) = n_j(2\omega)$*, it is said to be type I phase matching; when the relation is* $1/2\,(n_i(\omega) + n_j(\omega)) = n_j(2\omega)$*, it is type II phase-matching.*

5. **Second harmonic generation in a KDP crystal**

   (a) KDP is a negative uniaxial crystal ($n_o > n_e$). What is the polarisation of the wave at 2ω ?

   (b) Show that, in the case of type I phase-matching, the amplitude of the wave at 2ω satisfies the equation:

   $$\frac{\partial A_\theta(2\omega)}{\partial s} = \frac{i(2\omega)}{2nc} \chi^{(2)}_{eff,I} A_o^2(\omega),$$

   where $\chi^{(2)}_{eff,I}$ is an effective susceptibility, to be expressed as a funtion of elements of the tensor $\underline{\underline{\underline{\chi}}}^{(2)}(\omega,\omega)$.

   (c) Same question about type II phase-matching.

   (d) *Effective susceptibility calculation:* Using the following components of the nonlinear polarization in the coordinate system of the crystal:

   $$\begin{aligned} P_x(2\omega) &= 2\epsilon_o d_{14} E_y(\omega) E_z(\omega) \\ P_y(2\omega) &= 2\epsilon_o d_{14} E_x(\omega) E_z(\omega) \\ P_z(2\omega) &= 2\epsilon_o d_{36} E_x(\omega) E_y(\omega) \end{aligned}.$$

   Calculate the expressions of $\chi^{(2)}_{eff,I}$ and $\chi^{(2)}_{eff,II}$ as a function of the angles θ and ϕ.

   (e) *Optimization of second harmonic intensity* For each phase-matching type, determine how the crystal must be cut and how the incident wave must be polarized in order to optimize the second harmonic generation in the KDP crystal.