NONLINEAR OPTICS Tutorial – SHG

$\xrightarrow{\omega} \boxed{\chi^{(2)}} \rightarrow \omega, \rightarrow 2\omega$

$\vec{E}(\omega) = \vec{e}_i\, A(\omega)\, e^{i k_i(\omega) z}$

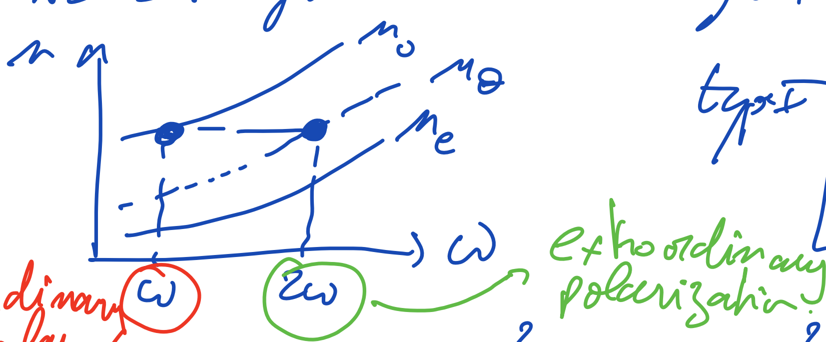
Type I $\Rightarrow$ $\vec{e}_i = \vec{e}_o$ (ord.) or $\vec{e}_\theta$ (extra)

1.1) Type I Phase matching condition

1 – $\vec{P}_{NL}(2\omega) = \varepsilon_0 \underline{\underline{\chi}}^{(2)}(2\omega;\omega,\omega)\, \vec{e}_i \vec{e}_i\, A^2(\omega)\, e^{2 i k_i(\omega) z}$

$\Rightarrow$ phase matching condition: $\boxed{2 k_i(\omega) = k(2\omega)}$

KDP = negative uniaxial crystal



ordinary polar.

type I phase matching

$\boxed{n_o(\omega) = n_\theta(2\omega)}$

2 – $\dfrac{1}{n_\theta^2(2\omega)} = \dfrac{\cos^2\theta}{n_o^2(2\omega)} + \dfrac{\sin^2\theta}{n_e^2(2\omega)} = \dfrac{1}{n_o^2(\omega)}$

$\Rightarrow \cos^2\theta \left[\dfrac{1}{n_o^2(2\omega)} + \dfrac{1}{n_e^2(2\omega)} \right] = \dfrac{1}{n_o^2(\omega)}$

$\Rightarrow \boxed{\theta = 41^\circ} \Rightarrow$ θ angle for which $\Delta k = 0$

1.2) Optimization of the nonlinear interaction

$\vec{E}(\omega) = \vec{e}_o\, A(\omega)\, e^{i k(\omega) z}$ and $\vec{E}(2\omega) = \vec{e}_\theta\, A(2\omega)\, e^{i k(2\omega) z}$

$$\Rightarrow \vec{P}_{NL}(2\omega) = \varepsilon_0 \underline{\underline{\chi}}^{(2)} \vec{e}_0 \vec{e}_0 A_\omega^2 e^{2ik(\omega)z}$$

with $\vec{e}_0 \left| \begin{array}{l} e_x = \sin\phi \\ e_y = -\cos\phi \\ e_z = 0 \end{array} \right.$

$$\Rightarrow \vec{P}_{NL}(2\omega) = \left| \begin{array}{l} P_x = \underline{\underline{2}} \varepsilon_0 \chi^{(2)}_{xyz} e_y e_z A^2(\omega) e^{2ik(\omega)z} \\ P_y = \underline{\underline{2}} \varepsilon_0 \chi^{(2)}_{yxz} e_x e_z \text{———} \\ P_z = 2\varepsilon_0 \chi^{(2)}_{zxy} e_x e_y \text{———} \end{array} \right.$$

$$\Rightarrow \boxed{P_{NL}(2\omega) = \left| \begin{array}{l} P_x = 0 \\ P_y = 0 \\ P_z = -\underline{\underline{2}} \varepsilon_0 \chi^{(2)}_{zxy} \sin\phi\cos\phi A^2(\omega) e^{2ik(\omega)z} \end{array} \right.}$$

2. $\vec{e}_\theta = \left| \begin{array}{l} -\cos\theta\cos\phi \\ -\cos\theta\sin\phi \\ \sin\theta \end{array} \right.$ $\quad \dfrac{\partial A_2}{\partial z} = \dfrac{i2\omega}{2nc\varepsilon_0} \vec{e}_2 \cdot \vec{P}_{NL}(2\omega) e^{-ik(2\omega)z}$

$$\Rightarrow \vec{e}_2 \cdot \vec{P}_{NL}(\omega) = \vec{e}_\theta \cdot \vec{P}_{NL}(2\omega)$$

$$= \varepsilon_0 \chi^{(2)} \sin 2\phi \sin\theta A_1^2$$

$$\Rightarrow \boxed{\frac{\partial A(2\omega)}{\partial z} = \frac{i2\omega}{2nc} \chi^{(2)}_{eff} A^2(\omega) \quad (\Delta k = 0)}$$

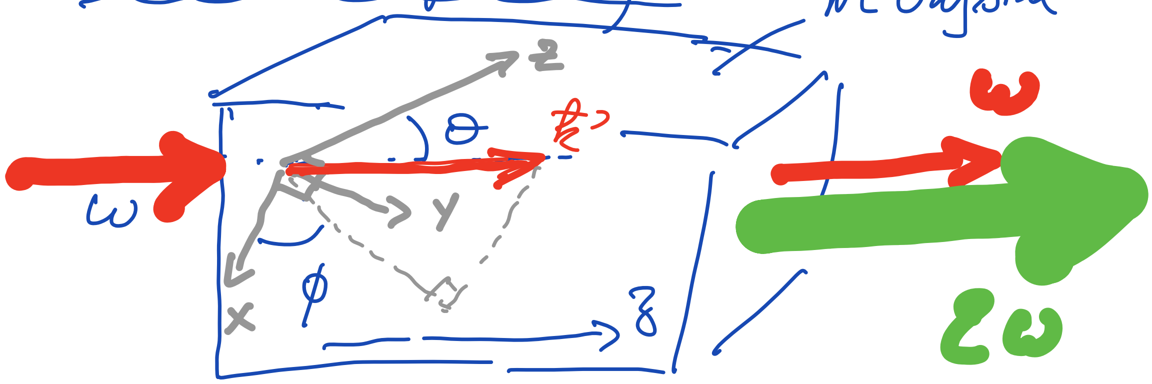
with $\boxed{\chi^{(2)}_{eff} = \chi^{(2)}_{zxy} \sin 2\phi \sin\theta}$

↳ Effective NL susceptibility (for Type I phase matching condition)

3 - $\chi^{(2)}_{eff}$ is maximized for $2\phi = \frac{\pi}{2} [\pi]$

$\Rightarrow \boxed{\phi = \frac{\pi}{4} [\pi/2]}$

1.3) Conversion efficiency



NL crystal cut

$$\frac{\partial A_2}{\partial z} = \frac{i\,2\omega}{2nc} \chi^{(2)}_{eff} A_1^2 \Rightarrow A_2(z) = A_2(0) + \frac{i\omega}{nc} \chi^{(2)}_{eff} A_1^2 z$$

Assuming $A_2(0) = 0$

$$\Rightarrow A_2(z) = \frac{i\omega}{nc} \chi^{(2)}_{eff} A_1^2(0)\, z$$

Now $I_2 = 2nc\varepsilon |A_2|^2$ (Intensity)

$$\rightarrow \boxed{I_2(\xi) = \frac{\omega^2}{n^2c^2}\left|\chi^{(2)}_{eff}\right|^2 \frac{I_1^2}{2nc\varepsilon_0}\xi^2}$$

SHG efficiency:

$$\boxed{\eta_0 = \frac{I_2(\xi)}{I_2(0)} = \frac{\omega^2}{2n^3c^3\varepsilon_0}\left|\chi^{(2)}_{eff}\right|^2 I_1 \xi^2} \quad (1)$$

under the undepleted pump approximation

2- $E = 1\,mJ$ Beam diameter $D = 1\,mm$
$\Delta\tau = 300\,ps$ crystal length $L = 10\,mm$.

$$I_1 = \frac{E}{\Delta\tau}\frac{1}{\pi D^2/4} \simeq 4\,10^{12}\,W/m^2$$

$$\Rightarrow \boxed{\eta_0 = 36\,\%}$$

3. The undepleted pump approximation is not fairly satisfied. For a more accurate evaluation, one needs to solve the coupled equations @ ω and 2ω.

1.4) Limit of the low conversion efficiency approx

Taking into account the pump depletion one finds: $\eta = \frac{I_{2\omega}(L)}{I_\omega(0)} = \tanh^2\left(\text{—}\right)$

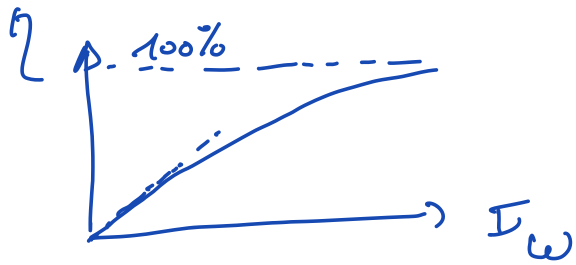
1. At low pump intensity

$$\eta = \tanh^2(x) \simeq x^2 = \underbrace{\frac{2\omega^2 |\chi_{eq}|^2 L^2}{\varepsilon_0 c\, n_{2\omega} n_\omega^2 \,]^2} I_\omega}_{= \eta_0}$$

we retrieve the relation (1)

At high intensity

$$\tanh(x) \xrightarrow{x \to \infty} 1 \Rightarrow \eta = 100\,\%$$



2. $$\eta = \frac{I_{2\omega}(L)}{I_\omega(0)} = \tanh^2\left(\sqrt{\eta_0}\right) = 28\,\%$$

to be compared with $\eta_0 = 36\,\%$ found in the case of undepleted pump approx.

2.1) Angular acceptance

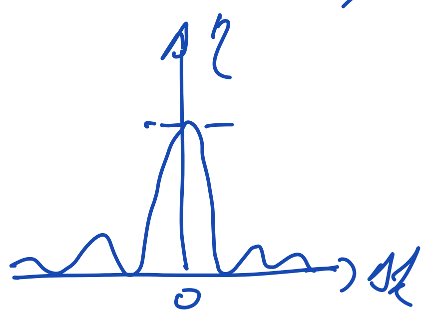
1. Solution of the nonlinear wave equation in case of phase mismatch:

$$\frac{\partial A_{2\omega}}{\partial z} = \frac{i\omega}{n_2 c} \chi^{(2)}_{eq} A_1^2 e^{i\Delta k z}$$

$$\Rightarrow A_{2\omega}(L) = \frac{i\omega}{n_2 c}\chi^{(2)}_{eff} A_1^2 \int_0^L e^{i\Delta k z}\,dz$$

$$\Rightarrow A_{2\omega}(L) = \frac{i\omega}{n_2 c}\chi^{(2)}_{eff} A_1^2 \,\mathrm{sinc}\left(\frac{\Delta k L}{2}\right)\frac{L}{2}\times\left(-i\, e^{i\frac{\Delta k}{2}L}\right)$$

$$\Rightarrow \boxed{\frac{I_{2\omega}(L)}{I_{\omega}(0)} \propto \left(\frac{\sin\frac{\Delta k L}{2}}{\frac{\Delta k L}{2}}\right)^2}$$



2- $\Delta k = \left[n_o(\omega) - n_\theta(\omega)\right]\frac{2\omega}{c}$

$$\Rightarrow \delta[\Delta k] = \frac{\partial \Delta k}{\partial\theta}\delta\theta = \frac{2\omega}{c}\frac{\partial n_\theta}{\partial\theta}\delta\theta$$

As $\frac{1}{n_\theta^2} = \frac{\cos^2\theta}{n_o^2} + \frac{\sin^2\theta}{n_e^2} \Rightarrow \frac{\partial n_\theta}{\partial\theta} = \dots$

$$\hookrightarrow \frac{\partial n_\theta}{\partial\theta} = \frac{\sin 2\theta}{2} n_\theta^3\left(\frac{n_e^2 - n_\theta^2}{n_o^2 n_e^2}\right)$$

$$= \frac{\sin 2\theta}{2} n_\theta^3 \frac{\overbrace{(n_e + n_\theta)}^{\simeq 2n_e}(n_e - n_\theta)}{n_o^2\, n_e^2}$$

$n_e \simeq n_o \simeq n_\theta$

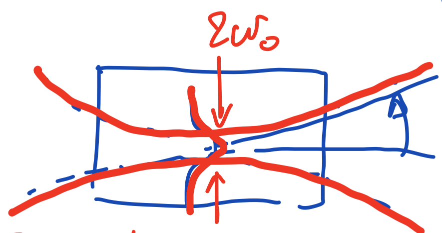
$$\Rightarrow \boxed{\frac{\partial n_\theta}{\partial\theta} \simeq (n_e - n_o)\sin 2\theta}$$

<u>Numerical Application:</u> $\frac{\Delta k L}{2} = \xi = 1{,}39 = \frac{2\omega}{c}\frac{\partial n_\theta}{\partial \theta}\,\delta\theta_{1/2}\,\frac{L}{2}$

$$\Rightarrow \boxed{\delta\theta_{1/2} = \frac{\xi\,\lambda_{pump}}{2\pi L\,(n_e - n_o)\sin 2\theta}}$$

$$\delta\theta_{1/2} = 5\,10^{-4}\ \text{rad} = 0{,}5\ \text{mrad}.$$

<u>Comments</u> $\Rightarrow$ link between the angular acceptance and the finite beam size.



Focused beam

$\delta\theta = \frac{\lambda}{n\pi w_0}$ = Beam divergence

inside the crystal

$\Rightarrow$ Constrainte : $\delta\theta < \delta\theta_{1/2}$

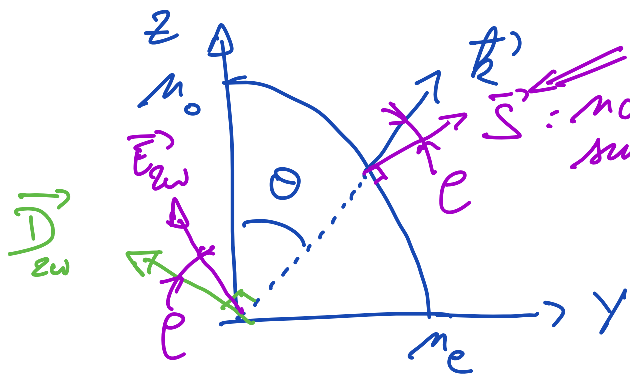
$$\Rightarrow \boxed{w_0 > \frac{\lambda}{n\pi\,\delta\theta_{1/2}}} \Rightarrow w_0 > 375\ \mu m$$

If the beam diameter is smaller (in order to increase the pump intensity for a higher efficiency), the beam divergence will exceed the angular acceptance $\delta\theta_{1/2}$ implying a reduction of the SHG efficiency.

3. For $\theta = \pi/2 \Rightarrow \sin 2\theta = 0 \Rightarrow \frac{\partial n}{\partial \theta} = 0$

$\Rightarrow$ Non critical phase matching condition.

2.2) Walk-off effect

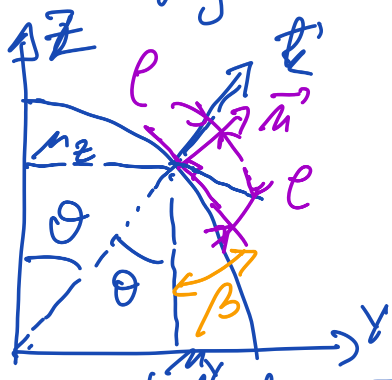


Poynting vector $\vec{S}$: normal to the surface of indices @ 2ω

ρ = walk-off angle.

The surface of indices defines : $\omega(\vec{k}) = c\,\frac{k}{n}$
$\Rightarrow$ The group velocity $\vec{v}_g = \frac{d\omega}{d\vec{k}}$ is given by the vector $\perp$ to the surface of indices
$\Rightarrow$ Now $\vec{v}_g \parallel \vec{S}$ the Poynting vector.

{ As the wave @ 2ω is extraordinary polarized, the walk-off angle $\rho \neq 0$ (see fig below)



Determination of ρ?

About the sign for the angles:

- $\theta \in [0, \pi/2] \Rightarrow \theta > 0$
- $\beta \in [0, \pi/2] \Rightarrow \beta > 0$
- The walk-off angle ρ is oriented from $\vec{k}$ to $\vec{S}$

{ positive crystal $\Rightarrow \rho > 0$
{ negative — $\Rightarrow \rho < 0$

$\Rightarrow \boxed{-\rho + \theta + \beta = \frac{\pi}{2}}$ where $\tan\beta = -\frac{dn_Y}{dn_Z}$

$$\Rightarrow \tan\beta = \tan\left(\frac{\pi}{2}+\rho-\theta\right)$$

$$= \frac{1}{\tan(\theta-\rho)} = -\frac{dn_r}{dn_z} \Rightarrow \boxed{\tan(\theta_c) = \frac{-dn_z}{dn_r}}$$

Surface of indices (negative uniaxial crystal case)

$$\frac{n_r^2}{n_e^2} + \frac{n_z^2}{n_o^2} = 1 \Rightarrow \frac{dn_z}{dn_r} = -\frac{n_o^2}{n_e^2}\,\frac{n_r}{n_z}$$

$$= -\frac{n_o^2}{n_e^2}\tan\theta$$

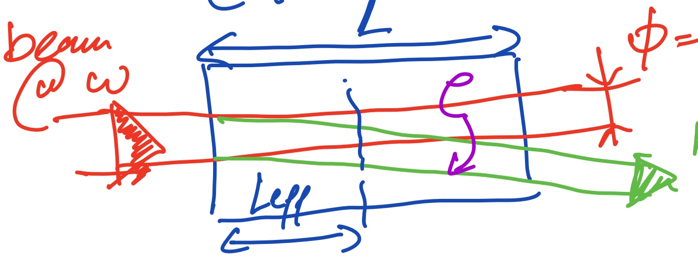
Now $\tan(\theta-\rho) = \dfrac{\tan\theta - \tan\rho}{1+\tan\theta\tan\rho} = \dfrac{n_o^2}{n_e^2}\tan\theta$

$$\Rightarrow \boxed{\tan\rho = \frac{1}{2}\, n_\theta^2 \left(\frac{n_e^2 - n_o^2}{n_o^2 n_e^2}\right)\sin 2\theta}$$

in the present case: $\boxed{\rho = 1.6^\circ}$

Comments:

- For a non critical phase matching situation
  $2\theta = \pi/2 \Rightarrow \tan\rho = 0 \quad \rho = 0$
- For $\rho \neq 0 \Rightarrow$ Reduction of the interaction length
  $\Rightarrow$ Effective length



$$\boxed{L_{eff} \simeq \frac{\phi}{\tan\rho}}$$

OK $\Leftarrow L_{eff} = 36$ mm (> 10 mm)