TUTORIAL Non linear Optics

3 Wave Mixing

1) Sum Freq generation in BBO

1- $\omega_3 = \omega_1 + \omega_2$ and $\omega_2 = 2\omega_1 \Rightarrow \lambda_3 = \frac{\lambda_1}{3} = 354$ nm.

2-
$$\left\{\begin{aligned}
\frac{dA_3}{dz} &= \frac{i\omega_3}{2n_3 c\varepsilon_0}\, \vec{e_3}\cdot \vec{P}_{NL}(\omega_3)\, e^{-ik_3 z} \\
\frac{dA_2}{dz} &= \frac{i\omega_2}{2n_2 c\varepsilon_0}\, \vec{e_2}\cdot \vec{P}_{NL}(\omega_2)\, e^{-ik_2 z} \\
\frac{dA_1}{dz} &= \frac{i\omega_1}{2n_1 c\varepsilon_0}\, \vec{e_1}\cdot \vec{P}_{NL}(\omega_1)\, e^{-ik_1 z}
\end{aligned}\right. \quad (1)$$

with
$$\vec{P}_{NL}(\omega_3) = \varepsilon_0\, \underline{\underline{\chi}}^{(2)}(\omega_3;\omega_1,\omega_2)\,\vec{E}(\omega_1)\,\vec{E}(\omega_2) + \varepsilon_0\, \underline{\underline{\chi}}^{(2)}(\omega_3;\omega_2,\omega_1)\,\vec{E}(\omega_1)\,\vec{E}(\omega_2)$$

$$\vec{P}_{NL}(\omega_{\frac{2}{1}}) = \varepsilon_0\, \underline{\underline{\chi}}^{(2)}(\omega_2;\omega_3,-\omega_{\frac{1}{2}})\,\vec{E}(\omega_3)\,\vec{E}(-\omega_{\frac{1}{2}}) + \varepsilon_0\, \underline{\underline{\chi}}^{(2)}(\omega_2;-\omega_{\frac{1}{2}},\omega_3)\,\vec{E}(-\omega_{\frac{1}{2}})\,\vec{E}(\omega_3)$$

$$(1) \Rightarrow \left\{\begin{aligned}
\frac{dA_3}{dz} &= \frac{i\omega_3}{2n_3 c}\,\chi^{(2)}_{eff,3}\, A_1 A_2\, e^{i(k_1+k_2-k_3)z} \\
\frac{dA_2}{dz} &= \frac{i\omega_2}{2n_2 c}\,\chi^{(2)}_{eff,2}\, A_3 A_1^*\, e^{i(k_3-k_1-k_2)z} \\
\frac{dA_1}{dz} &= \frac{i\omega_1}{2n_1 c}\,\chi^{(2)}_{eff,1}\, A_3 A_2^*\, e^{i(k_3-k_2-k_1)z}
\end{aligned}\right.$$

with the effective susceptibilities:

$$\left\{\begin{aligned}
\chi^{(2)}_{eff,3} &= \vec{e_3}\cdot\underline{\underline{\chi}}^{(2)}(\omega_3;\omega_1,\omega_2)\,\vec{e_1}\,\vec{e_2} + \vec{e_3}\cdot\underline{\underline{\chi}}^{(2)}(\omega_3;\omega_2,\omega_1)\,\vec{e_1}\,\vec{e_2} \\
&= 2\,\vec{e_3}\cdot\underline{\underline{\chi}}^{(2)}(\omega_3;\omega_1,\omega_2)\,\vec{e_1}\,\vec{e_2} \\
\chi^{(2)}_{eff,\frac{2}{1}} &= \vec{e_{\frac{2}{1}}}\cdot\underline{\underline{\chi}}^{(2)}(\omega_{\frac{2}{1}};\omega_3,-\omega_{\frac{1}{2}})\,\vec{e_3}\,\vec{e_{\frac{1}{2}}} + \vec{e_{\frac{2}{1}}}\cdot\underline{\underline{\chi}}^{(2)}(\omega_{\frac{2}{1}};-\omega_{\frac{1}{2}},\omega_3)\,\vec{e_{\frac{1}{2}}}\,\vec{e_3} \\
&= 2\,\vec{e_{\frac{2}{1}}}\cdot\underline{\underline{\chi}}^{(2)}(\omega_{\frac{2}{1}};\omega_3,-\omega_{\frac{1}{2}})\,\vec{e_3}\,\vec{e_{\frac{1}{2}}}
\end{aligned}\right.$$

3) Phase matching condition $k_1 + k_2 = k_3$

4) $I_i = 2 n_i c \varepsilon_0 |A_i|^2$

$$\Rightarrow \frac{dI_i}{dz} = 2 n_i c \varepsilon_0 \left[A_i \frac{dA_i^*}{dz} + \frac{dA_i}{dz} A_i^* \right]$$

$$\Rightarrow \frac{dI_3}{dz} = +i \varepsilon_0 \omega_3 \chi^{(2)}_{eff,3} A_1 A_2 A_3^* e^{i\Delta k z} + cc$$

$$(2) \left| \begin{aligned} \frac{dI_2}{dz} &= -i \varepsilon_0 \omega_2 \chi^{(2)*}_{eff,2} A_1 A_2 A_3^* e^{i\Delta k z} + cc \\ \frac{dI_1}{dz} &= -i \varepsilon_0 \omega_1 \chi^{(2)*}_{eff,1} A_1 A_2 A_3^* e^{i\Delta k z} + cc \end{aligned} \right.$$

5) Energy conservation : $\frac{dI_1}{dz} + \frac{dI_2}{dz} + \frac{dI_3}{dz} = 0$

$\Rightarrow$ Now from (2), we get $\frac{dI_1}{dz} + \frac{dI_2}{dz} + \frac{dI_3}{z} = \omega_3 \chi^{(2)}_{eff,3} - \omega_2 \chi^{(2)}_{eff,2} - \omega_1 \chi^{(2)}_{eff,1}$

So the necessary condition to fulfill is : $\boxed{\chi^{(2)}_{eff,3} = \chi^{(2)*}_{eff,2} = \chi^{(2)*}_{eff,1}}$

$\hookrightarrow \chi^{(2)}_{eff}$ should'd be a real quantity

$\hookrightarrow$ Dispersion of $\chi^{(2)}$ with frequency is neglegible

6) $\Delta k = 0$

$$\Rightarrow \frac{dA_3}{dz} = \frac{i\omega_3}{2 n_3 c} \chi^{(2)}_{eff} A_1 A_2$$

It means that

$$\cancel{\frac{dN_1}{dz} + 2\frac{dN_2}{dz} + 3\frac{dN_3}{dz} = 0}$$

$$\boxed{\frac{dN_3}{dz} = -\frac{dN_2}{dz} = -\frac{dN_1}{dz}}$$

COMMENT

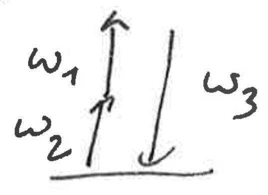
From (2) we can get:

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

as $I = N \hbar \omega$

$\hookrightarrow N$: nbr of photon $/ m^2$

~~and as $\omega_3 = 3\omega_1$, $\omega_2 = 2\omega_1$~~



7) $\Delta k = 0$

$$\frac{dA_3}{dz} = \frac{3i\omega}{2nc}\chi_m^{(2)} A_1 A_2$$

$$\Rightarrow A_3(L) = \underbrace{A_3(0)}_{=0} + \frac{3i\omega}{n_3 c}\chi_m^{(2)} A_1 A_2 L$$

$$\Rightarrow I_3(L) = \frac{9\omega^2}{n_3^2 c^2}|\chi_m^{(2)}|^2 \frac{I_2}{2n_2 c\varepsilon_0}\frac{I_1}{\cancel{2n_1 c\varepsilon_0}} L\, \cancel{2n_3 c\varepsilon_0}$$

$$\boxed{I_3(L) = \frac{9\omega^2}{2n^3 c^3\varepsilon_0}|\chi_m^{(2)}|^2 I_2 I_1 L}$$

8) $L = 10\ mm$

$R = 200\ \mu m$

$\chi_m^{(2)} = 4\ pm/V$

$$E_{nergy} = P_{peak} \times \Delta z \Rightarrow I = \frac{P_{peak}}{\pi\frac{\phi^2}{4}} = \frac{4E}{\Delta z\,\pi R^2}$$

$I_1 = 40\ GW/cm^2$

$I_2 = 8\ GW/cm^2$

$I_3 = 6\ GW/cm^2$

$\Rightarrow E(3\omega) = 8\ \mu J/pulse \Rightarrow$ undepleted approximation is limit

Tutorial Nonlinear Optics

3 wave Mixing

2) Second Harmonic generation

1- Wave equation @ 2ω : $\frac{\partial A(2\omega)}{\partial z} = \frac{i2\omega}{2nc\varepsilon_0} \vec{e}_{\omega}^{\,*} \cdot \vec{P}_{nl}(2\omega)\, e^{-ik_{2\omega}z}$

$$\vec{P}_{nl}(2\omega) = \varepsilon_0\, \underline{\underline{\chi}}^{(2)}(2\omega,\omega,\omega)\, \vec{e}_\omega \vec{e}_\omega\, A(\omega) A(\omega)\, e^{2ik(\omega)z}$$

$$\Rightarrow \boxed{\frac{\partial A(2\omega)}{\partial z} = \frac{i2\omega}{2nc}\, \chi_{nl}^{(2)} A^2(\omega)\, e^{-i\Delta k z}}$$ with $\Delta k = k_{2\omega} - 2k(\omega)$

and $\chi_{nl}^{(2)} = \vec{e}_{2\omega}^{\,*} \cdot \underline{\underline{\chi}}^{(2)} \vec{e}_\omega \vec{e}_\omega$

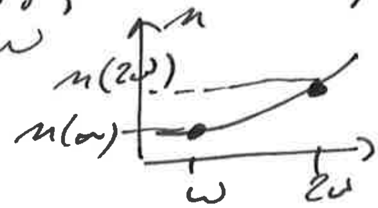
So the phase matching term = $\boxed{\Delta k = k_{2\omega} - 2k_\omega}$

Phase matching condition?

$\Delta k = 0 \Rightarrow k_{2\omega} = 2k_\omega$ for collinear interaction.

$\Rightarrow n(2\omega)\frac{2\omega}{c} = 2\, n(\omega)\frac{\omega}{c} \Rightarrow n(2\omega) = n(\omega)$ IMPOSSIBLE

The phase matching is generally impossible to satisfy as the refractive index increases with ω



and it is verified in our case as

$n_\omega = 2.1555$

$n_{2\omega} = 2.234.$

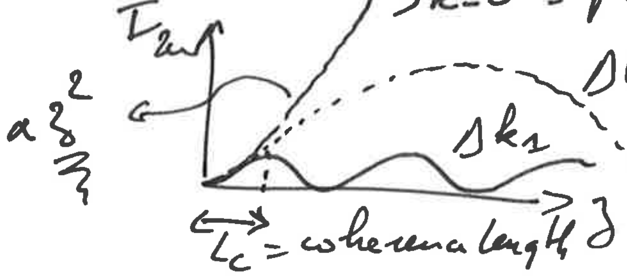
2- $A_{2\omega}(z) = \frac{i2\omega}{2nc}\chi_{nl}^{(2)} A_\omega^2 \int_0^z e^{-i\Delta k z}\, dz$

assuming a nondepleted pump : $A_\omega(z) = cste$

$$A_{2\omega}(z) = \frac{i2\omega}{2nc}\chi_{nl}^{(2)} A_\omega^2\, \frac{2}{\Delta k} \sin\left(\frac{\Delta k}{2} z\right) e^{-i\Delta k z}$$

As $I = 2nc\varepsilon|A|^2$

$$\Rightarrow \boxed{I_{2\omega}(z) = \frac{(2\omega)^2}{2n^3c^3\varepsilon_0}\,|\chi_{nl}^{(2)}|^2 \sin^2\left(\frac{\Delta k}{2} z\right) \frac{I_\omega^2}{\Delta k^2}}$$



$\Delta k = 0$ = perfect phase matching.

For $\Delta k \neq 0$, sinusoidal variation of the intensity with $z \Rightarrow$ pure NL efficiency.

· For $\Delta k = 0$: $I_{2\omega} = \frac{(2\omega)^2}{2n^3c^3\varepsilon_0} |\chi^{(2)}_{eff}|^2 I_\omega^2 L^2$

as $I_{2\omega}(z)$ can be expressed in terms of sinc

$$I_{2\omega}(z) \propto \text{sinc}^2\left(\frac{\Delta k}{2} z\right) I_\omega^2 z^2$$

3 - For $\Delta k \neq 0$, the max efficiency is reached for:

$$\frac{\Delta k z}{2} = \frac{\pi}{2} \Rightarrow \boxed{z = \frac{\pi}{\Delta k}}$$ coherence length.

Example $LiNbO_3$ case

$$\Delta k = \frac{2\omega}{c}\left[n(2\omega) - n(\omega)\right] \Rightarrow L_{coh} = \frac{\pi}{\Delta k} = 3,8\,\mu m \text{ !!}$$

meaning that the interaction length is reduced to few μm !!

$$\eta_{SHG} = \frac{I_{2\omega}}{I_\omega} = \text{SHG efficiency} = \frac{(2\omega)^2}{2n^3c^3\varepsilon_0} |\chi^{(2)}_{eff}|^2 \frac{I_\omega}{(\pi/L_{coh})^2}$$

$$\eta_{SHG} = \frac{(2\omega)^2}{2n^3c^3\varepsilon_0} |\chi^{(2)}_{eff}|^2 \frac{I_\omega}{\pi^2} L_{coh}^2 = 4.6\,10^{-7} \text{ !!}$$

$(I_\omega = 4,5\ MW/cm^2)$ $\Downarrow$ very weak efficiency

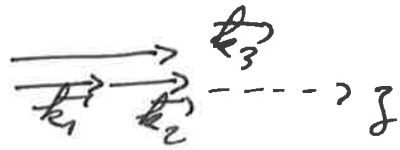
4 - $\Delta k = 0$: $\eta_{SHG} = \frac{(2\omega)^2}{8n^3c^3\varepsilon_0} |\chi^{(2)}_{eff}|^2 I_\omega z^2$

↳ under the undepleted pump approximation

$\Rightarrow$ for $z = 2mm \Rightarrow \eta_{SHG} = 31\%$ meaning that the approximation on the pump depletion is not fully correct.

3- Sum freq. generation - SUPPLEMENT

The phase matching condition in a colinear interaction is given by: $k_3 = k_1 + k_2$



↳ actually this condition is derived from a more general vectorial relation $\vec{k_3} = \vec{k_1} + \vec{k_2}$

↳ as $\vec{k}_{3,2,1} = k_{3,2,1}\,\vec{z} \Rightarrow k_3 = k_1 + k_2$ along z

$$k_3 = k_1 + k_2 \Rightarrow n_3 \frac{\omega_3}{c} = n_1 \frac{\omega_1}{c} + n_2 \frac{\omega_2}{c}$$

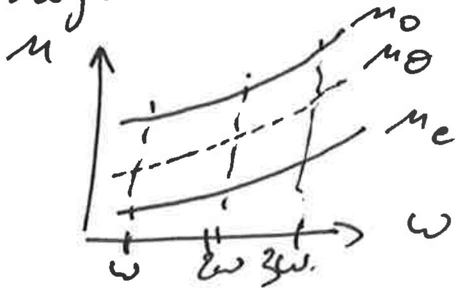
as $\omega_3 = 3\omega_1$, $\omega_2 = 2\omega_1 \Rightarrow$ $\boxed{3 n_3 = n_1 + 2 n_2}$ (1)

The interacted waves are either polarized along the ordinary or the extraordinary direction
e_o $\vec{e_\theta}$

In order to satisfy the relation (1), necessarly we have

$n_1 < n_3 < n_2$ or $n_2 < n_3 < n_1$

BBO = negative uniaxial crystal ($n_o > n_e$)



type I phase matching

$$3\, n_{\theta}(3\omega) = n_o(\omega) + 2\, n_o(2\omega)$$

type II phase matching

$$3\, n_{\theta}(3\omega) = n_\theta(\omega) + 2\, n_o(2\omega)$$

or

$$3\, n_{\theta}(3\omega) = n_o(\omega) + 2\, n_\theta(2\omega)$$

BBO case

type I: $n_\theta(3\omega) = \dfrac{n_o(\omega) + 2 n_o(2\omega)}{3} = 1.6676$

↳ The value is comprised between $n_o(3\omega)$ and $n_e(3\omega)$. So it is possible to find an angle θ to reach $n(3\omega) = 1.6676$ for which $\Delta k = 0$.