2021 Nov 10

NLO #10

- Acousto-optic modulation

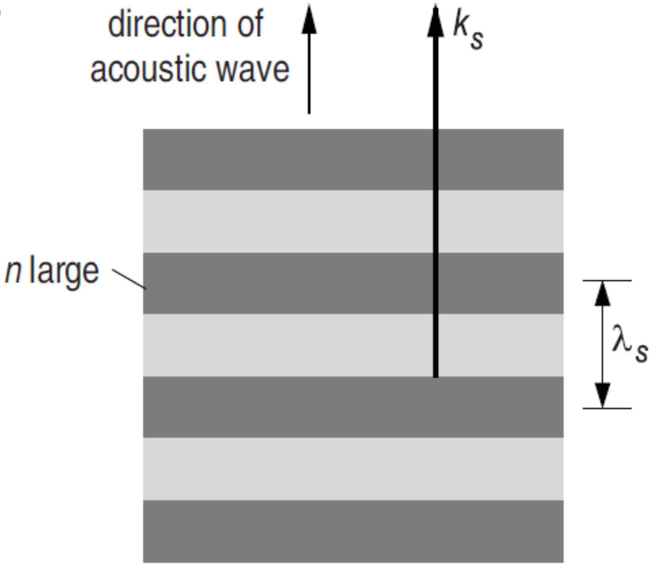
- CHi3-Effects

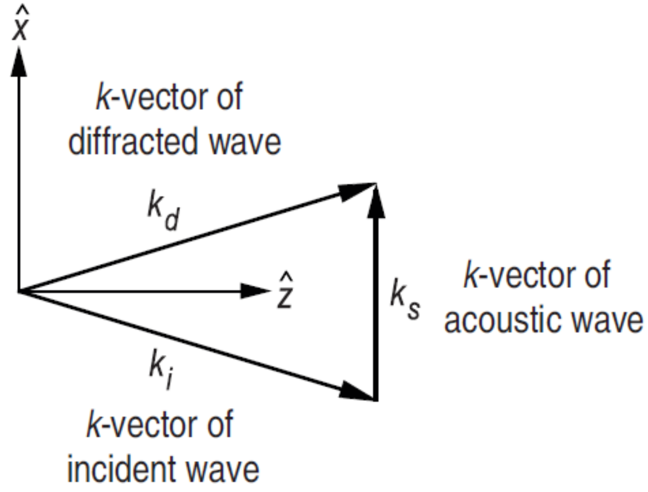
- chi3 – Tensor
- chi3 – Processes
  - THG
  - SPM
  - XPM
  - FWM
  - Self-focusing / Self-trapping
  - SPM for pulses

Acousto-optic modulators (travelling wave)

$$\nabla \times \nabla \times \mathbf{E} = -\mu_0 \frac{\partial^2}{\partial t^2}\mathbf{D}$$

$$\begin{aligned}\mathbf{D} &= \varepsilon_0 \mathbf{E} + \mathbf{P} = \varepsilon_0 \left(n + \Delta n(r,t)\right)^2 \mathbf{E} \\ &\approx \varepsilon_0 n^2 \mathbf{E} + 2\varepsilon_0 n \underline{\Delta n(r,t)\mathbf{E}},\end{aligned}$$





Acoustic wave:

$$\begin{aligned}\Delta n(r,t) &= \Delta \hat{n} \cos\left(\omega_s t - \mathbf{k}_s \cdot \mathbf{r}\right) \\ &= \frac{\Delta \hat{n}}{2}\left[e^{j(\omega_s t - \mathbf{k}_s\cdot\mathbf{r})} + e^{-j(\omega_s t - \mathbf{k}_s\cdot\mathbf{r})}\right]\end{aligned}$$

Optical wave:

$$\mathbf{E} = \hat{\mathbf{E}}_i e^{j(\omega_i t - \mathbf{k}_i\cdot\mathbf{r})} + \underline{\hat{\mathbf{E}}_d e^{j(\omega_d t - \mathbf{k}_d\cdot\mathbf{r})} + c.c.}$$

$$\begin{aligned}\mathbf{k}_d &= \mathbf{k}_s + \mathbf{k}_i \\ \omega_d &= \omega_s + \omega_i\end{aligned}$$

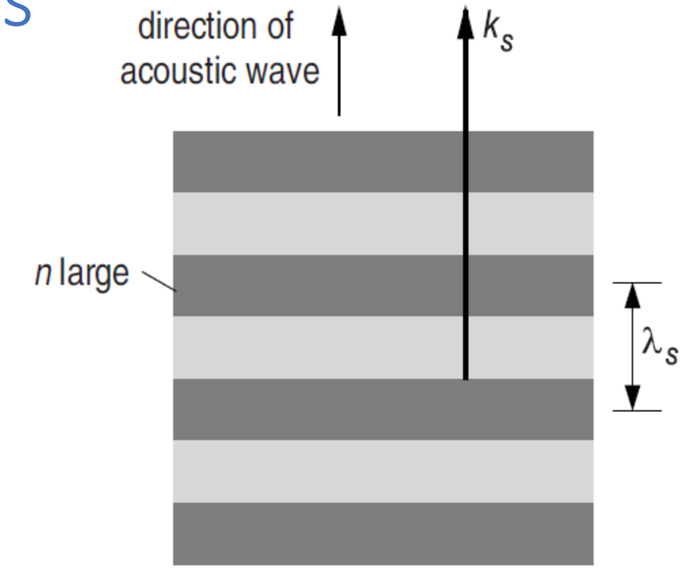
$$\nabla \times \nabla \times \mathbf{E} + \frac{1}{c^2}\frac{\partial^2 \mathbf{E}}{\partial t^2} = -2\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\left[\frac{\Delta n(r,t)}{n}\mathbf{E}\right]$$

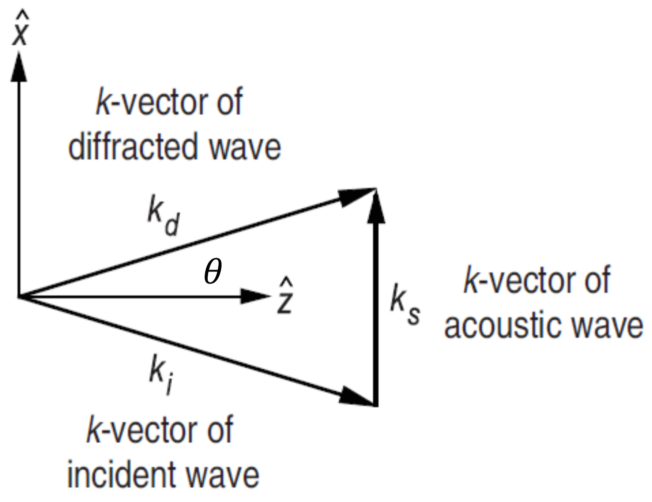
$$\mathbf{E}\cdot\nabla\varepsilon = 0$$

if E-field is y-polarized

$$\mathbf{\Delta E} - \frac{1}{c^2}\frac{\partial^2 \mathbf{E}}{\partial t^2} = +2\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\left[\frac{\Delta n(r,t)}{n}\mathbf{E}\right]$$

Acousto-optic modulators (travelling wave)





$$
\begin{aligned}
\mathbf{E}_i &= \mathbf{e}_y A_i(z) e^{j(\omega_i t - \mathbf{k}_i \cdot \mathbf{r})} + c.c. \\
\mathbf{E}_d &= \mathbf{e}_y A_d(z) e^{j(\omega_d t - \mathbf{k}_d \cdot \mathbf{r})} + c.c.
\end{aligned}
$$

Insert fields in wave equation and apply SVEA:

$$
\begin{aligned}
-\cancel{\left(\mathbf{k}_d^2 - \frac{\omega_d^2}{c^2}\right)} A_d(z) - 2j\mathbf{k}_d \cdot \nabla A_d(z) &\simeq -\frac{\omega_d^2}{c^2} \Delta\hat{n} A_i(z) \\
-\cancel{\left(\mathbf{k}_i^2 - \frac{\omega_i^2}{c^2}\right)} A_i(z) - 2j\mathbf{k}_i \cdot \nabla A_i(z) &\simeq -\frac{\omega_i^2}{c^2} \Delta\hat{n} A_d(z)
\end{aligned}
\quad
\xrightarrow[|\mathbf{k}_d| \simeq |\mathbf{k}_i|]{\mathbf{k}_d \cdot \nabla A_d(z) = k_d \cos\theta \frac{dA_d}{dz}}
\quad
\begin{aligned}
\frac{dA_d(z)}{dz} &\simeq -j \frac{\omega_d}{2c} \frac{\Delta\hat{n}}{\cos\theta} A_i(z) \\
\frac{dA_i(z)}{dz} &\simeq -j \frac{\omega_i}{2c} \frac{\Delta\hat{n}}{\cos\theta} A_d(z)
\end{aligned}
$$

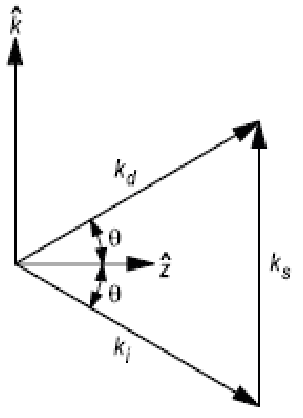
$$
\boxed{
\begin{aligned}
A_i(z) &= A_i(0) \cos|\kappa|\, z \\
A_d(z) &= -jA_i(0) \sin|\kappa|\, z
\end{aligned}
}
\qquad
\kappa = \frac{\omega_d}{2c} \frac{\Delta\hat{n}}{\cos\theta} \simeq \frac{\omega_i}{2c} \frac{\Delta\hat{n}}{\cos\theta}
$$

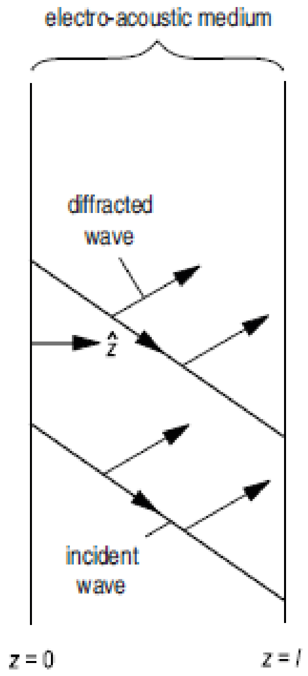
- Beam steering
- Frequency shifting

Acousto-optic modulators (standing wave)

Standing acoustic wave:

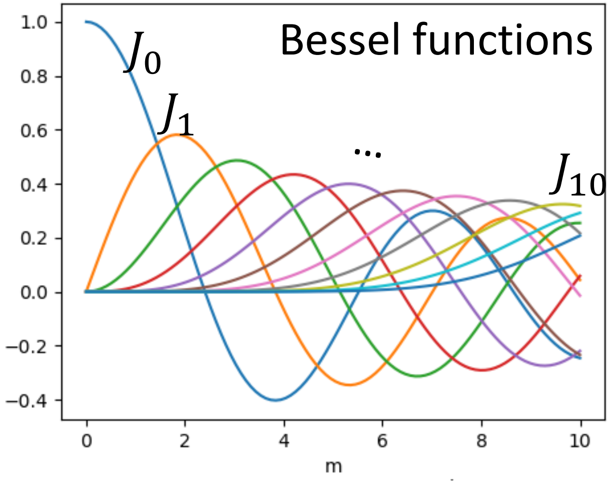
$$\begin{aligned}\Delta n(\mathbf{r},t) \;&=\; \Delta n \sin\omega_s t \cos(\mathbf{k}_s\mathbf{r}) = \frac{\Delta n}{4j}\{\exp[j(\omega_s t - \mathbf{k}_s\mathbf{r})] \\ &\quad + \exp[j(\omega_s t + \mathbf{k}_s\mathbf{r})] - \exp[-j(\omega_s t - \mathbf{k}_s\mathbf{r})] \\ &\quad - \exp[-j\omega_s t + \mathbf{k}_s\mathbf{r}]\}\,.\end{aligned}$$





$\omega_d = \omega_i \pm m\omega_s$ (phase matched for up to large m)

$$\begin{aligned}A_i(\ell) &= A_i(0)\cos\left(\frac{\omega_i}{c}\frac{\Delta n}{2\cos\theta}\ell\right) \\ A_d(\ell) &= -jA_i(0)\sin\left(\frac{\omega_i}{c}\frac{\Delta n}{2\cos\theta}\ell\right)\end{aligned}
\quad \xrightarrow{\Delta n(t) = \Delta n \sin\omega_s t} \quad
\begin{aligned}A_i(\ell) &= A_i(0)\sum_{m\,\text{even}} J_m\left(\frac{\omega_i}{c}\frac{\Delta n}{2\cos\theta}\ell\right)e^{jm\omega_s t}, \\ A_d(\ell) &= -A_i(0)\sum_{m\,\text{odd}} J_m\left(\frac{\omega_i}{c}\frac{\Delta n}{2\cos\theta}\ell\right)e^{jm\omega_s t}\end{aligned}$$



Amplitude modulation (of $m = 0$ zero order beam)

Third-order susceptibility and symmetries

$$\chi^{(3)}_{ijkl}$$

3x3x3x3 = 81 elements (accounting for polarization)
also depends on involved optical frequencies

$$P_i = \ldots + \epsilon_0 \chi^{(3)}_{ijkl} E_j E_k E_l + \ldots$$

(sum over double indices)

This greatly simplifies for **<u>isotropic</u>** materials:
(nothing should change when rotating, inverting, mirroring the material)

$$\chi_{1111} = \chi_{2222} = \chi_{3333},$$
$$\chi_{1122} = \chi_{1133} = \chi_{2211} = \chi_{2233} = \chi_{3311} = \chi_{3322},$$
$$\chi_{1212} = \chi_{1313} = \chi_{2323} = \chi_{2121} = \chi_{3131} = \chi_{3232},$$
$$\chi_{1221} = \chi_{1331} = \chi_{2112} = \chi_{2332} = \chi_{3113} = \chi_{3223}.$$

All other elements with odd number of occurrence of one index are zero

Moreover:

$$\chi_{1111} = \chi_{1122} + \chi_{1212} + \chi_{1221}$$

(only 3 independent elements)

If **<u>Kleinman's symmetry</u>** is assumed
(lossless, all frequencies well below material resonance):

- Frequency independence
- instantaneous
- Order of indices does not matter

$$\chi_{1111} = 3\,\chi_{1122}$$

Overview $\chi^{(3)}$ processes

$$\frac{\partial^2}{\partial z^2}E - \epsilon_0\epsilon_r\mu_0\frac{\partial^2}{\partial t^2}E = \mu_0\frac{\partial^2}{\partial t^2}P_{NL}$$

assume only one polarization

$$P_{\mathrm{NL}} = \epsilon_0\chi^{(3)}E^3$$

$$E(z,t) = \sum_n E_n(z)\mathrm{e}^{-i(\omega_n t - k_n z)} + \mathrm{c.c.}$$

index indicates frequency

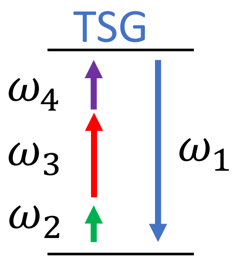
$$\Downarrow \quad |k_n E_n| >> \left|\frac{\partial}{\partial z}E_n\right|$$

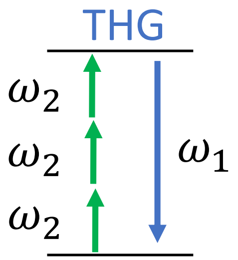
$$\sum_m 2ik_m\frac{\partial}{\partial z}E_m\mathrm{e}^{-i(\omega_m t - k_m z)} = \mu_0\epsilon_0\chi^{(3)}\sum_{n,p,q}(\omega_n+\omega_p+\omega_q)^2E_nE_pE_q\mathrm{e}^{-i[(\omega_n+\omega_p+\omega_q)t-(k_n+k_p+k_q)z]}$$
$$+ 3\mu_0\epsilon_0\chi^{(3)}\sum_{n,p,q}(\omega_n+\omega_p-\omega_q)^2E_nE_pE_q^*\mathrm{e}^{-i[(\omega_n+\omega_p-\omega_q)t-(k_n+k_p-k_q)z]}$$

Require that this equation holds for each frequency component separately:

$$\boxed{\begin{aligned}\frac{\partial}{\partial z}E_m = &-i\frac{\omega_m}{2cn_m}\chi^{(3)}\sum_{n,p,q}\delta(\omega_n+\omega_p+\omega_q-\omega_m)E_nE_pE_q\mathrm{e}^{i(k_n+k_p+k_q-k_m)z}\\ &-i\frac{3\omega_m}{2cn_m}\chi^{(3)}\sum_{n,p,q}\delta(\omega_n+\omega_p-\omega_q-\omega_m)E_nE_pE_q^*\mathrm{e}^{i[(k_n+k_p-k_q-k_m)z}\end{aligned}}$$

Overview $\chi^{(3)}$ processes



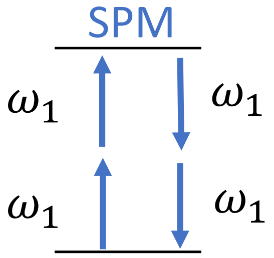


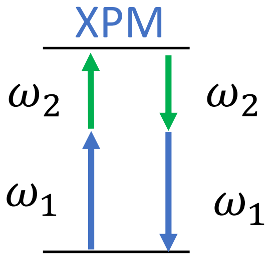
Triple-sum (TSG)
Third harmonic (THG)

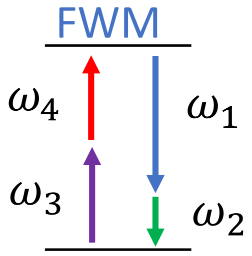
$$\frac{\partial}{\partial z}E_m = -i\frac{\omega_m}{2cn_m}\chi^{(3)}\sum_{n,p,q}\delta\underline{(\omega_n+\omega_p+\omega_q-\omega_m)}E_nE_pE_q\mathrm{e}^{i(k_n+k_p+k_q-k_m)z}$$

$$-i\frac{3\omega_m}{2cn_m}\chi^{(3)}\sum_{n,p,q}\delta\underline{(\omega_n+\omega_p-\omega_q-\omega_m)}E_nE_pE_q^*\mathrm{e}^{i[(k_n+k_p-k_q-k_m)z}$$

Self-phase modulation (SPM)
Cross-phase modulation (XPM)
Four-wave mixing (FWM)

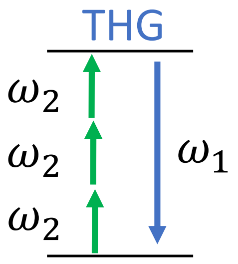






Third-harmonic generation

Two frequencies:
$$\omega_m = 3\omega_n$$



$$\frac{\partial}{\partial z}E_m = -\,i\frac{\omega_m}{2cn_m}\chi^{(3)}\sum_{n,p,q}\delta(\omega_n+\omega_p+\omega_q-\omega_m)E_nE_pE_q\mathrm{e}^{i(k_n+k_p+k_q-k_m)z}$$
$$-\,i\frac{3\omega_m}{2cn_m}\chi^{(3)}\sum_{n,p,q}\delta(\omega_n+\omega_p-\omega_q-\omega_m)E_nE_pE_q^*\mathrm{e}^{i[(k_n+k_p-k_q-k_m)z}$$

$$\frac{\partial}{\partial z}E_m = -i\frac{\omega_m}{2cn_m}\chi^{(3)}E_nE_nE_n\mathrm{e}^{i\Delta kz} \qquad \Delta k = 3k_n - k_m$$

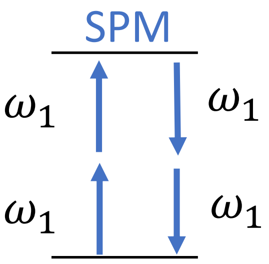
Solution:

$$E_m(l) = -i\frac{\omega_m}{2cn_m}\chi^{(3)}E_n^3\int_0^l \mathrm{e}^{i\Delta kz}\,\mathrm{d}z$$
$$= -i\frac{\omega_m}{2cn_m}\chi^{(3)}E_n^3\frac{\mathrm{e}^{i\Delta l}-1}{i\Delta k}$$

$$\boxed{\begin{aligned} I_m &= \left(\frac{\omega_m\chi^{(3)}}{2cn_m}\right)^2\left(\frac{1}{2cn_n\epsilon_0}\right)^2|I_n|^3l^2\left(\frac{\sin(\Delta kl/2)}{\Delta kl/2}\right)^2 \\ &= \left(\frac{\omega_m\chi^{(3)}}{8c^2n_mn_n\epsilon_0}\right)^2|I_n|^3l^2\,\mathrm{sinc}^2(\Delta kl/2)\end{aligned}}$$

Self-Phase modulation (SPM)

Only one frequency
ω_m



$$\frac{\partial}{\partial z}E_m = -i\frac{\omega_m}{2cn_m}\chi^{(3)}\sum_{n,p,q}\delta(\omega_n+\omega_p+\omega_q-\omega_m)E_nE_pE_q e^{i(k_n+k_p+k_q-k_m)z}$$
$$-i\frac{3\omega_m}{2cn_m}\chi^{(3)}\sum_{n,p,q}\delta(\omega_n+\omega_p-\omega_q-\omega_m)E_nE_pE_q^* e^{i[(k_n+k_p-k_q-k_m)z}$$

$$\frac{\partial}{\partial z}E_m = -i\frac{3\omega_m}{2cn_m}\chi^{(3)}(E_mE_m^*)E_m$$

Perfectly phase matched

Solution:

$$\boxed{E_m(z) = E_m(0)\exp\left[-i\underbrace{\frac{3\omega_m\chi^{(3)}}{2cn_m}|E_m|^2}_{\Delta k_m} z\right]}$$

$$\Delta n_m = \frac{3\chi^{(3)}}{2n_m}|E|^2$$

$$\boxed{\Delta n_m = \frac{3\chi^{(3)}}{4c\epsilon_0 n_m^2}I_m = n_2 I_m}$$

Intensity:
$I_m = 2n_m\epsilon_0 c|E_m|^2$

<u>Alternatively:</u>

$$(n_m + n_2 I_m)^2 = 1 + \chi_{\mathrm{eff,m}}$$
$$= 1 + \chi^{(1)} + 3\chi^{(3)}|E_m|^2$$
$$= 1 + \chi^{(1)} + \frac{3\chi^{(3)}}{2\epsilon_0 c n_m}I_m$$

$$n_2 = \frac{3\chi^{(3)}}{4\epsilon_0 c n_m^2}I_m$$

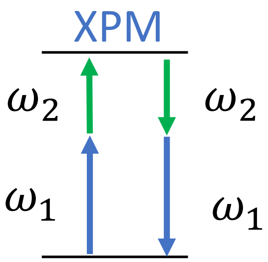
n_2 is the **<u>nonlinear refractive index</u>**
(---> intensity dependent refractive index)

$$\boxed{n_2 = \frac{3\chi^{(3)}}{4\epsilon_0 c n^2}I \qquad n = n + n_2 I}$$

Cross-Phase modulation (XPM)

Two frequencies

ω_m, ω_n



$$\frac{\partial}{\partial z}E_m = -i\frac{\omega_m}{2cn_m}\chi^{(3)}\sum_{n,p,q}\delta(\omega_n+\omega_p+\omega_q-\omega_m)E_nE_pE_q\mathrm{e}^{i(k_n+k_p+k_q-k_m)z}$$
$$-i\frac{3\omega_m}{2cn_m}\chi^{(3)}\sum_{n,p,q}\delta(\omega_n+\omega_p-\omega_q-\omega_m)E_nE_pE_q^*\mathrm{e}^{i[(k_n+k_p-k_q-k_m)z}$$

$$\frac{\partial}{\partial z}E_m = -i\frac{3\omega_m}{2cn_m}\chi^{(3)}(E_mE_m^*)E_m - \underline{2}\,i\frac{3\omega_m}{2cn_m}\chi^{(3)}(E_nE_n^*)E_m$$

Solution:

$$\boxed{E_m(z) = E_m(0)\exp\left[-i\left(\frac{3\omega_1\chi^{(3)}}{2cn_m}|E_m|^2 + \underline{2}\frac{3\omega_1\chi^{(3)}}{2cn_m}|E_n|^2\right)z\right]}$$

$$\boxed{\Delta n_m = n_2 I_m + \underline{2}n_2 I_n}$$

XPM twice as strong as SPM

$$\chi_{1111} = 3\,\chi_{1122}$$

In case of crossed polarizations, $\chi^{(3)}$ is reduced by factor of 3

SPM/XPM for circular polarized light

What happens if we have an arbitrary polarization:

$$\boldsymbol{E}(z) = E_x(z)\, \mathrm{e}^{-i(\omega t - kz)} \boldsymbol{e}_x + E_y(z)\, \mathrm{e}^{-i(\omega t - kz)} \boldsymbol{e}_y$$

Polarization

$$P_x = \epsilon_0 \chi^3 \left[3|E_x|^2 E_x + \frac{2 \cdot 3}{3} |E_y|^2 E_x + E_y^2 E_x^* \right]$$

$$P_y = \epsilon_0 \chi^3 \left[3|E_y|^2 E_y + \frac{2 \cdot 3}{3} |E_x|^2 E_y + E_x^2 E_y^* \right]$$

Circular pol. basis

$$\boldsymbol{\sigma}_\pm = \frac{\boldsymbol{e}_x \pm i \boldsymbol{e}_y}{\sqrt{2}}$$

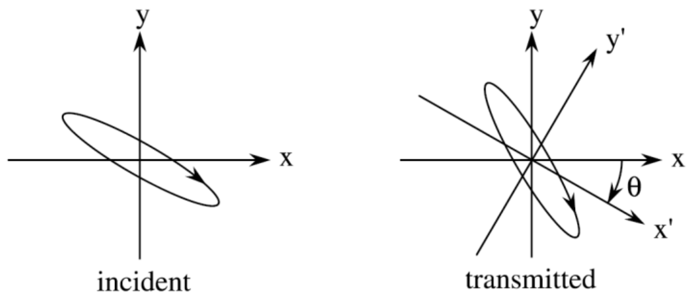
$$\boldsymbol{\sigma}_\pm^* = \boldsymbol{\sigma}_\mp$$

$$\boldsymbol{\sigma}_\pm \cdot \boldsymbol{\sigma}_\pm = 0$$

$$\boldsymbol{\sigma}_\pm \cdot \boldsymbol{\sigma}_\mp = 1$$

$$\boxed{P_\pm = 2\epsilon_0 \chi^{(3)} \left[|E_\pm|^2 E_\pm + 2|E_\mp|^2 E_\pm \right]}$$

For elliptically polarized light the polarization ellipse rotates

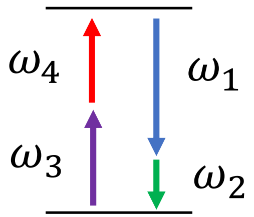


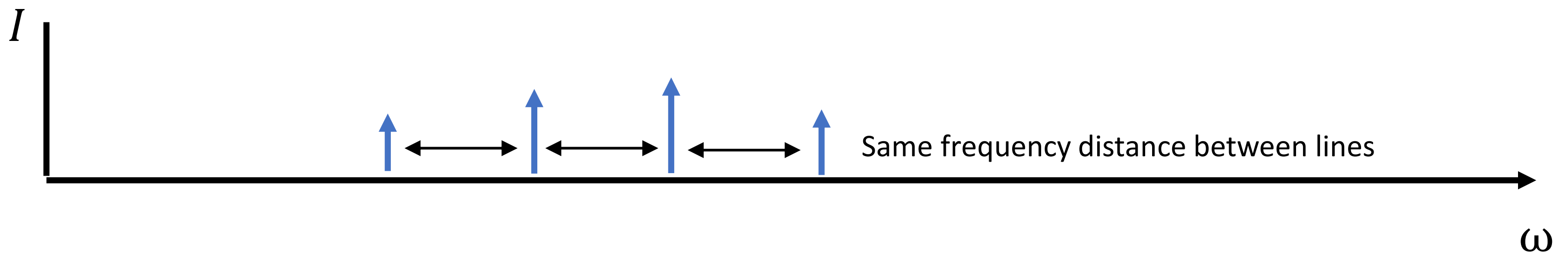
$$\boxed{n_2^{\mathrm{circ}} = \frac{2}{3} n_2^{\mathrm{lin}}}$$

- Circular polarized light remains circular
- XPM twice as strong as SPM

Four wave mixing (FWM)

- Photon number is conserved
- ‘Center of gravity’ of the spectrum is conserved
- Short range mixing (in terms of frequency separation)
- **SPM / XPM are important**

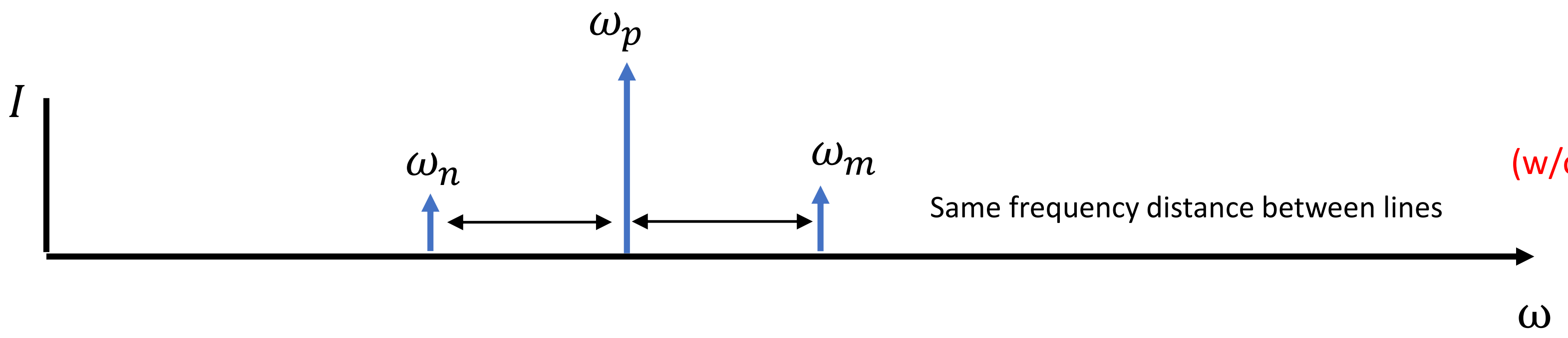




$$\frac{\partial}{\partial z}E_m = -i\frac{3\omega_m}{2cn_m}\chi^{(3)}\sum_{n,p,q}\delta(\omega_n+\omega_p-\omega_q-\omega_m)E_nE_pE_q^*\mathrm{e}^{i[(k_n+k_p-k_q-k_m)z}$$

(w/o SPM/XPM)

Parametric Amplifier

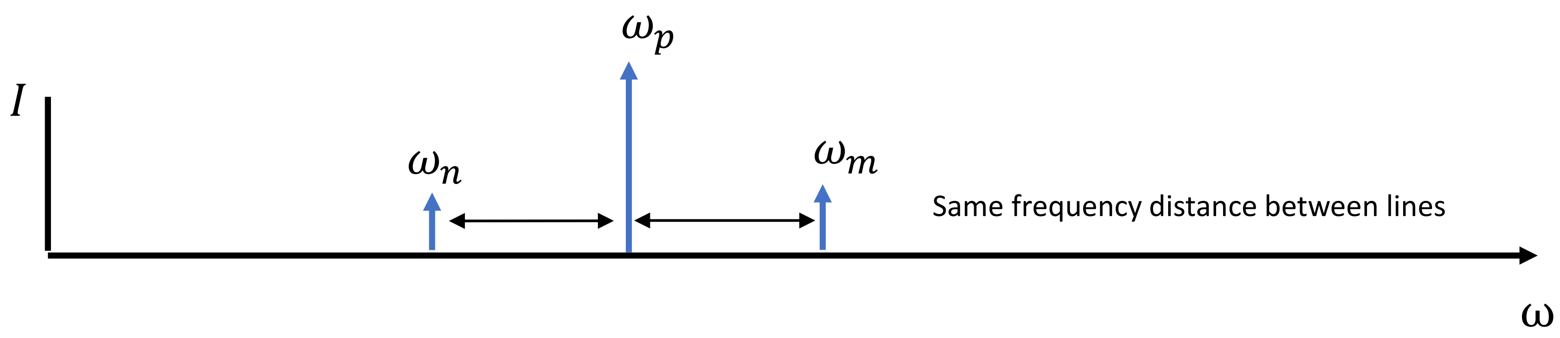


(w/o SPM/XPM)

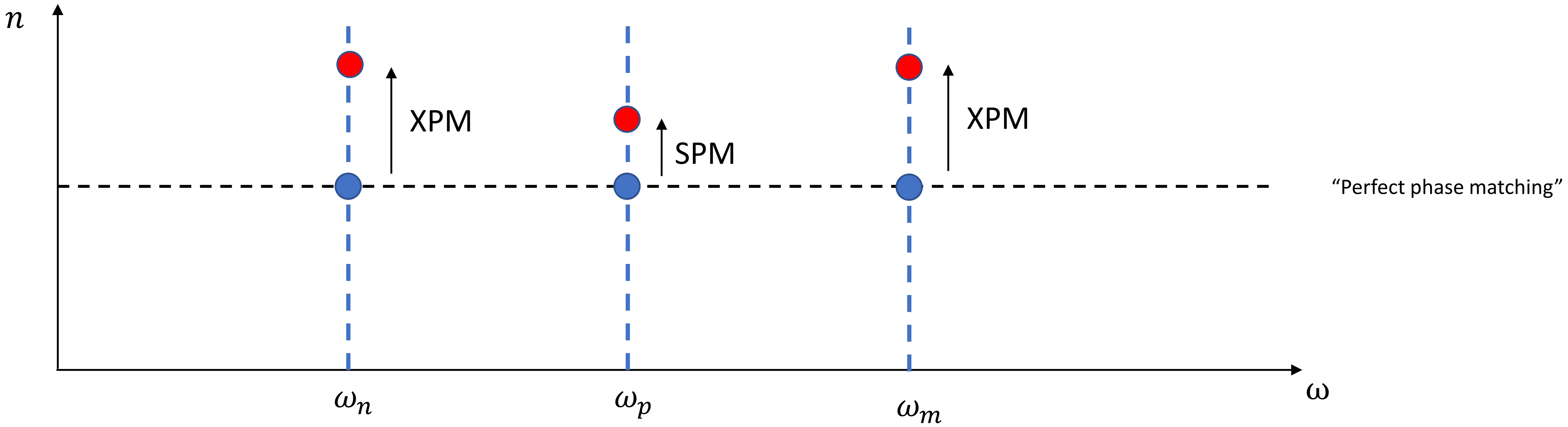
$$\frac{\partial}{\partial z}E_m = -i\frac{3\omega_m}{2cn_m}\chi^{(3)}\delta(\omega_p+\omega_p-\omega_n-\omega_m)E_pE_pE_n^*\mathrm{e}^{i[(2k_p-k_n-k_m)z}$$

Example: Importance of SPM/XPM and Dispersion

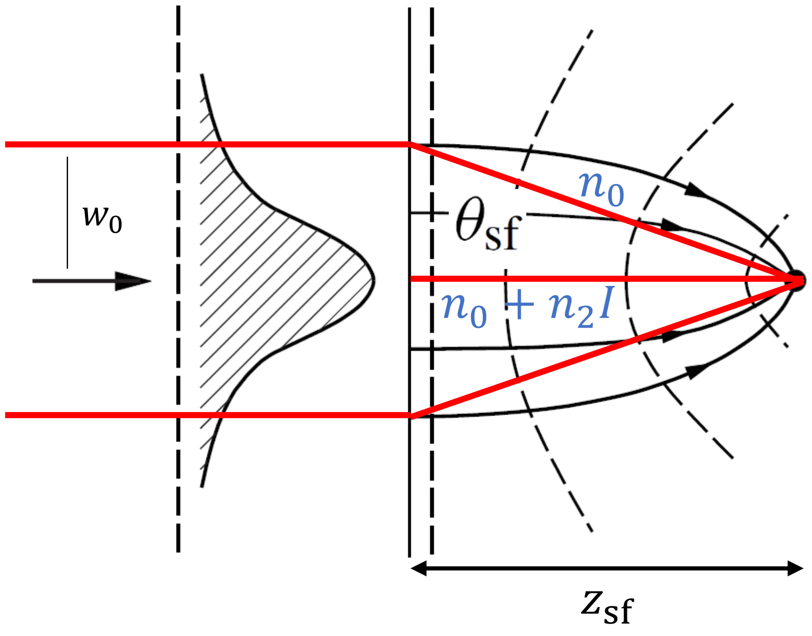
Parametric Amplifier



$$\frac{\partial}{\partial z}E_m = -i\frac{3\omega_m}{2cn_m}\chi^{(3)}\delta(\omega_p+\omega_p-\omega_n-\omega_m)E_pE_pE_n^*\mathrm{e}^{i[(2k_p-k_n-k_m)z}$$



Self-focusing and self-trapping



$$(n_0 + n_2 I) z_{sf} = n_0 z_{sf} / \cos\theta_{sf}$$

Self-focusing angle

$$\theta_{sf} = \sqrt{2 n_2 I / n_0}$$

Self-focusing distance

$$z_{sf} = w_0 \sqrt{\frac{n_0}{2 n_2 I}}$$

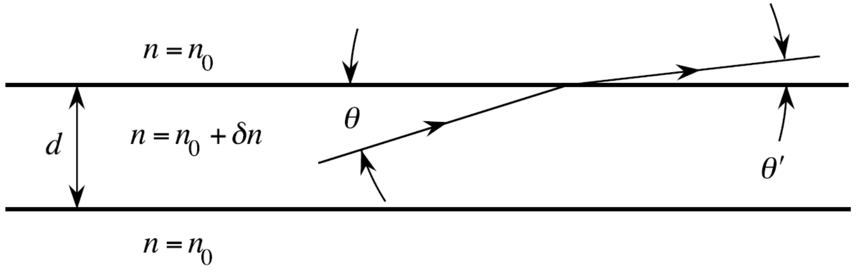
$\theta_{dif} = \theta_{sf}$

Diffraction angle of Gauss beam:

$$\theta_{dif} = \frac{\lambda}{\pi n w_0}$$

$$\boxed{P_{cr} \approx \frac{\lambda^2}{8 n_0 n_2}}$$

Power is relevant, *not* intensity

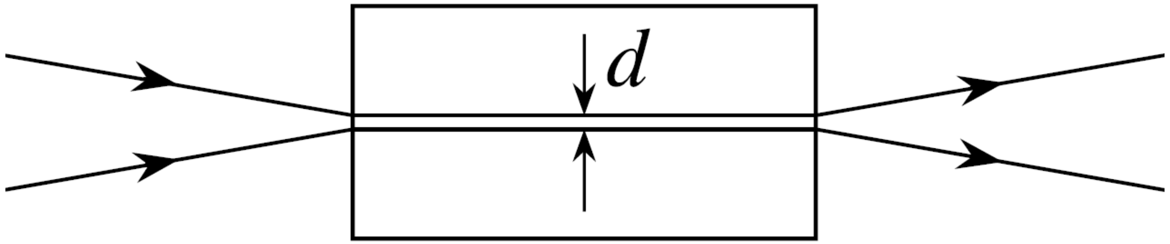


Limit angle for total internal reflection

$$\cos\theta_0 = \frac{n_0}{n_0 + \delta n} \qquad \theta_0 = (2\delta n / n_0)^{1/2}$$

$\theta_{dif} = \theta_0$

Spatial solitons



Balance between

$$2ik_0\frac{\partial A}{\partial z} + \nabla_T^2 A = -\frac{\omega^2}{\epsilon_0 c^2} p_{\mathrm{NL}} \qquad p_{\mathrm{NL}} = 3\epsilon_0 \chi^{(3)} |A|^2 A$$

$$2ik_0\frac{\partial A}{\partial z} + \frac{\partial^2 A}{\partial x^2} = -3\chi^{(3)}\frac{\omega^2}{c^2}|A|^2 A$$

Spatial soliton

$$A(x,z) = A_0 \operatorname{sech}(x/x_0) e^{i\gamma z}$$

$$x_0 = \frac{1}{k_0}\sqrt{\frac{n_0}{2\bar{n}_2 |A_0|^2}} = \frac{1}{k_0}\sqrt{\frac{n_0}{n_2 I}} \qquad \gamma = k_0 \bar{n}_2 |A_0|^2 / n_0 = k_0 n_2 I/(2n_0)$$