Third-order nonlinear effects

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Outline

- Coupled-wave equations for 3rd-order processes
- Third-harmonic generation (SHG)
- Phase-matching considerations
- Self-focusing & soliton formation
- Polarization effects in nonlinear optics
- Kerr electro-optical effect

Coupled-wave equations for 4-wave mixing

$$\frac{\partial \mathscr{E}_s}{\partial z} - \frac{i}{2k(\omega_s)}\nabla_\perp^2 \mathscr{E}_s = \frac{i\omega_s^2}{2k(\omega_s)c^2} \times \chi_{eff}^{(3)}(-\omega_s;\omega_1,\omega_2,\omega_3)\mathscr{E}_1\mathscr{E}_2\mathscr{E}_3 e^{i\Delta kz} \quad (1)$$

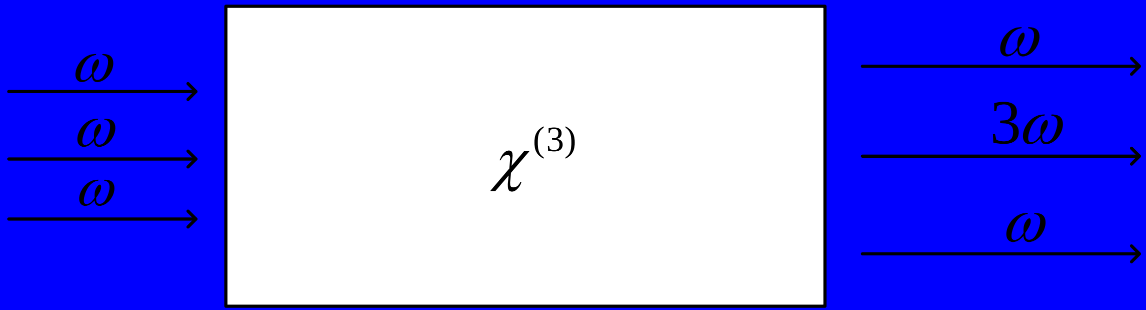
Effective nonlinear susceptibility:

$$\chi_{eff}^{(3)}(-\omega_s;\omega_1,\omega_2,\omega_3) \equiv c^{(3)}(\omega_1,\omega_2,\omega_3) \times \sum_{ijkl} \tilde{\chi}_{ijkl}^{(3)}(-\omega_s;\omega_1,\omega_2,\omega_3)\mathbf{e}_i(\omega_s)\mathbf{e}_j(\omega_1)\mathbf{e}_k(\omega_2)\mathbf{e}_l(\omega_3) \quad (2)$$

Phase mismatch:

$$\Delta k \equiv k(\omega_1) + k(\omega_2) + k(\omega_3) - k(\omega_s)$$

Application: Third-harmonic generation



$$\Delta k = 3k(\omega) - k(3\omega)$$

Coupled-wave equations in plane-wave goemetry:

$$\frac{d\mathscr{E}_\omega}{dz} = \frac{3i\omega}{2n_\omega c}\chi^{(3)}_{eff}\mathscr{E}_{3\omega}\mathscr{E}^{*2}_\omega e^{-i\Delta kz}$$

$$\frac{d\mathscr{E}_{3\omega}}{dz} = \frac{3i\omega}{2n_{3\omega} c}\chi^{(3)}_{eff}\mathscr{E}^{3}_\omega e^{i\Delta kz}$$

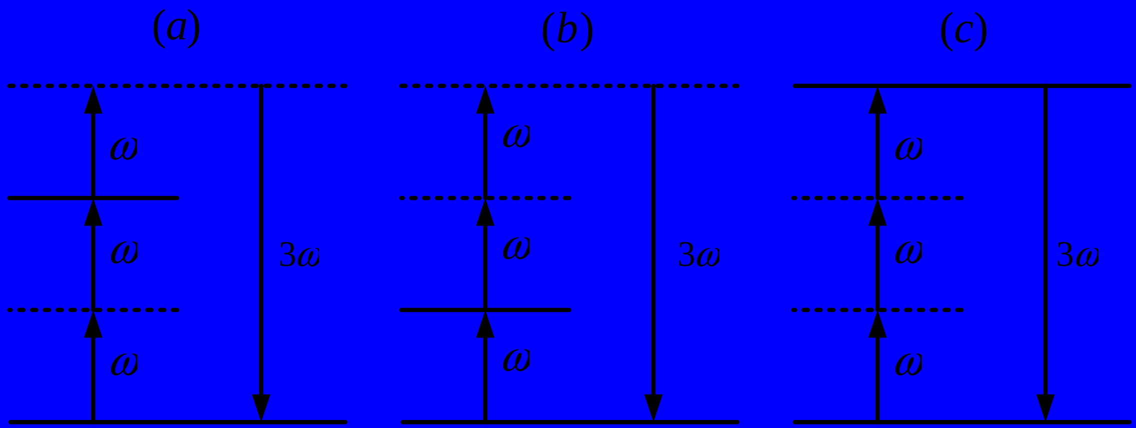
Efficiency estimate in solids:

- Input intensity $I \sim 100$ MW/cm^2 (dielectric breakdown in solids);
- crystal length $L \sim 1$ cm;
- nonlinearity: $\chi^{(3)}_{eff} \sim 10^{-21}$ m^2/W^2

$$\eta_{THG} \sim 5 \times 10^{-7} \ll 1, \Longrightarrow \text{no way!}$$

Can be realized in gases as a two-stage process:

$$FW \Longrightarrow SH \Longrightarrow TH$$

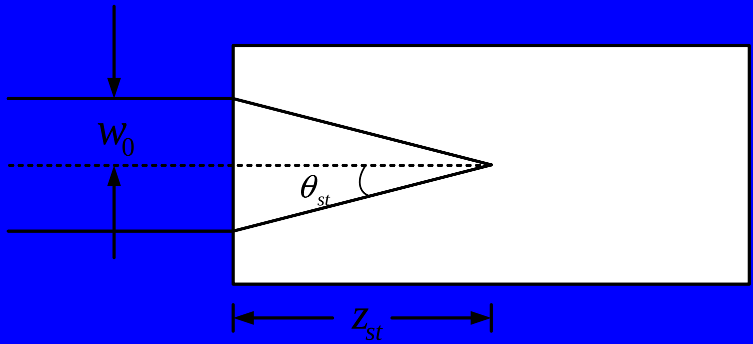


Self-focusing & spatial soliton formation

Beam propagation in a $\chi^{(3)}$-medium

- Diffraction of a beam of size w_0
- Self-focusing nonlinearity,

$$\Delta n_{NL} \sim \overline{n}_2 I_0 > 0$$



Diffraction length:

$$L_D \simeq k w_0^2,$$

Nonlinear length:

$$k \Delta n_{NL} L_{NL} \sim 1$$

implying

$$L_{NL} \sim \frac{1}{k \bar{n}_2 I_0}$$

- Balance of the two effects $\Longrightarrow$ optical soliton
- Optical solitons $\Longrightarrow L_{NL} = L_D$

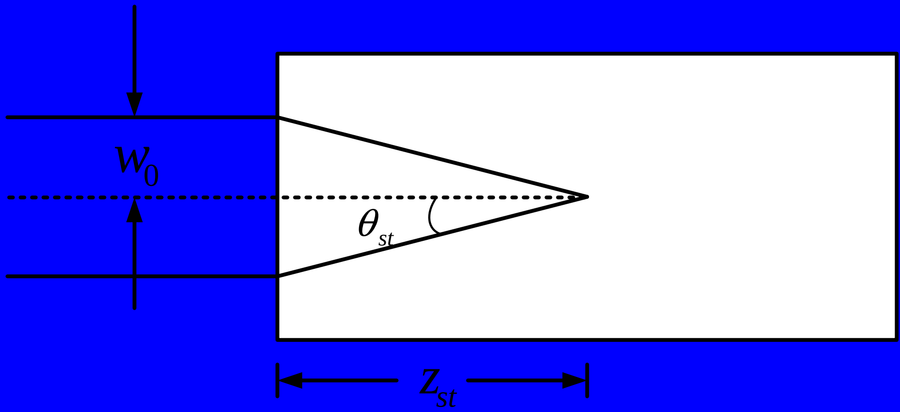
- Critical power for soliton formation:

$$P_{cr} \simeq \frac{\lambda_0^2}{4\pi n_0 \bar{n}_2}$$

where

$$\lambda_0 = 2\pi/k_0 = 2\pi c/\omega$$

- $L_D \ll L_{NL}$, linear regime, diffraction dominates;
- $L_{NL} \ll L_D$, self-phase modulation dominates



Estimating the focusing length:

$$z_f \simeq \frac{L_D}{2}\sqrt{\frac{P_{cr} n_0}{P}}$$

with

$$P \gg P_{cr}$$

Mathematical description of self-focusing

$$\frac{\partial \mathscr{E}_\omega}{\partial z} - \frac{i}{2k_\omega}\nabla_\perp^2 \mathscr{E}_\omega = \frac{i\omega^2}{2k_\omega c^2}\chi_{eff}^{(3)}(-\omega;\omega,-\omega,\omega) \times |\mathscr{E}_\omega|^2 \mathscr{E}_\omega \tag{3}$$

$$\chi_{eff}^{(3)} \equiv \frac{3}{4}\sum_{ijkl}\tilde{\chi}_{ijkl}^{(3)}(-\omega;\omega,-\omega,\omega) \times \mathbf{e}_i(\omega)\mathbf{e}_j(\omega)\mathbf{e}_k(\omega)\mathbf{e}_l(\omega) \tag{4}$$

Self-phase modulation regime

Introduce linear and nonlinear losses:

$$|\mathscr{E}_\omega|^2 = |\tilde{\mathscr{E}}_\omega|^2 e^{-\alpha(\omega)z}$$

and

$$\chi^{(3)}(\omega) = \chi_r^{(3)}(\omega) + i\chi_i^{(3)}(\omega)$$

$$\frac{\partial \tilde{\mathscr{E}}_\omega}{\partial z} = \frac{i\omega^2}{2k_\omega c^2}\chi^{(3)}(\omega)|\tilde{\mathscr{E}}_\omega|^2 \tilde{\mathscr{E}}_\omega e^{-\alpha(\omega)z}$$

$$\tilde{\mathscr{E}}_\omega = |\tilde{\mathscr{E}}_\omega| e^{i\Phi_\omega}$$

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Intensity evolution

$$I(\rho, z) = \frac{I_0(\rho)e^{-\alpha z}}{1 + \beta_2 I_0(\rho) L_{\text{eff}}(z)}$$

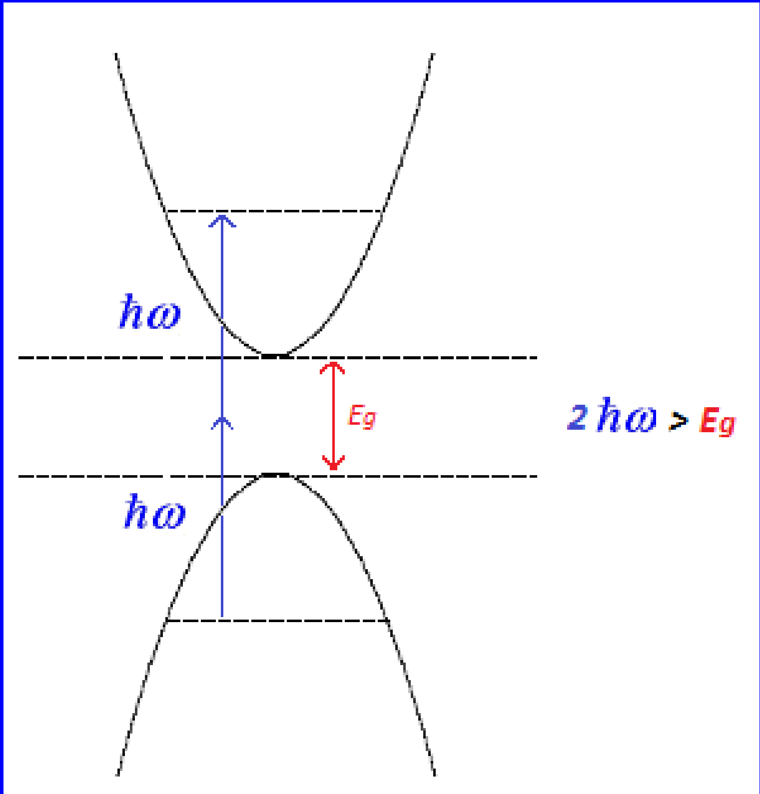
Effective interaction length:

$$L_{\text{eff}} = \frac{1}{\alpha}(1 - e^{-\alpha z})$$

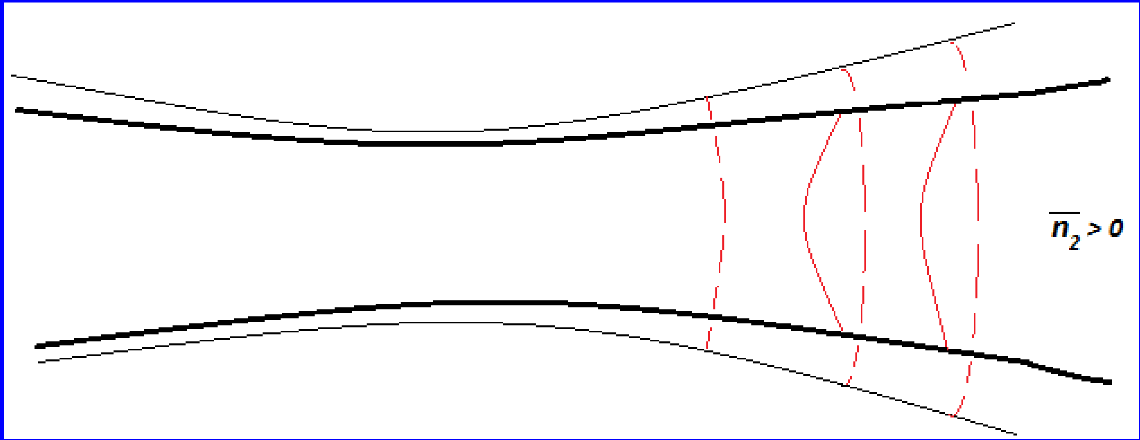
Two-photon absorption coefficient:

$$\beta_2 = \frac{3k_0\chi_i^{(3)}}{2\varepsilon_0 n_0^2 c}$$

Two-photon absorption in semiconductors



Phase Evolution: Nonlinear lens



$$\Delta\Phi(\rho) \simeq -\frac{k_0 \overline{n}_2 I_0 \rho^2}{2w_0^2}\Delta L$$

Assume linearly polarized beam:

$$P_{NL} = \frac{3}{4}\varepsilon_0 \chi^{(3)} |\mathscr{E}|^2 \mathscr{E} = \varepsilon_0 \chi_{NL} \mathscr{E}$$

Total refractive index:

$$n_{tot} = n_L + n_{NL},$$

Nonlinear refractive index

$$n_{NL} = n_2|\mathscr{E}|^2 = \bar{n}_2 I.$$

Optical intensity:

$$I = \frac{\varepsilon_0 c n_0}{2}|\mathscr{E}|^2$$

Connection between two nonlinear coefficients

$$n_2[m^2/V^2] = \frac{\varepsilon_0 c n_0}{2}\bar{n}_2[m^2/W]$$

Soliton units:

- Propagation distance: $Z = z/L_D$;
- Spatial coordinate: $X = x/w_0$;
- Soliton order parameter:

$$\mathcal{N}^2 \equiv \frac{L_D}{L_{NL}} = \frac{P}{P_{\mathrm{cr}}}$$

Dimensionless wave equation

$$i\frac{\partial U}{\partial Z} + \frac{1}{2}\frac{\partial^2 U}{\partial X^2} + \mathcal{N}^2 |U|^2 U = 0$$

- $\mathcal{N} = 1 \iff P = P_{\rm cr}$, fundamental soliton:

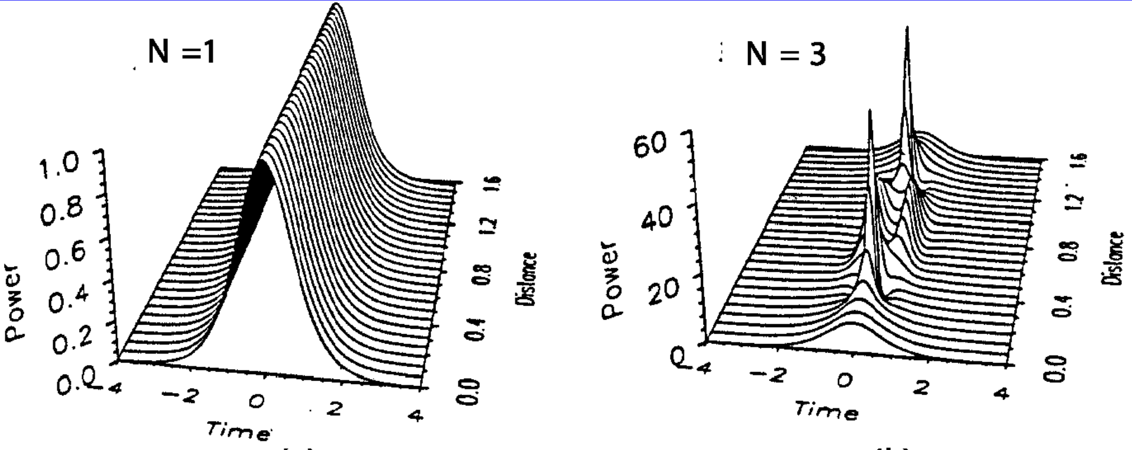
$$U(Z,X) = \operatorname{sech} X\, e^{-iZ/2}$$

Profile does not change on propagation!

- Space-time analogy $\Longrightarrow$ temporal solitons
- Propagation distance $Z = z/L_{dis}$, $L_{dis} = T_{\rm p}^2/\beta_2$;
- Time: $T = t/T_{\rm p}$

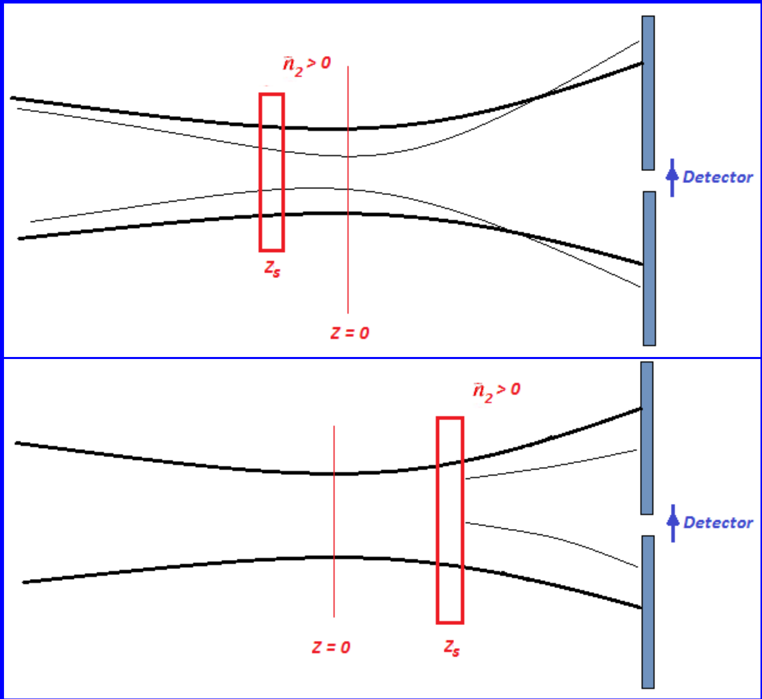
$$i\frac{\partial U}{\partial Z} + \frac{1}{2}\frac{\partial^2 U}{\partial T^2} + \mathcal{N}^2|U|^2 U = 0$$

- Fundamental soliton: $U(Z,T) = \text{sech}\, T\, e^{-iZ/2}$;
- Higher-order solitons (breathers):

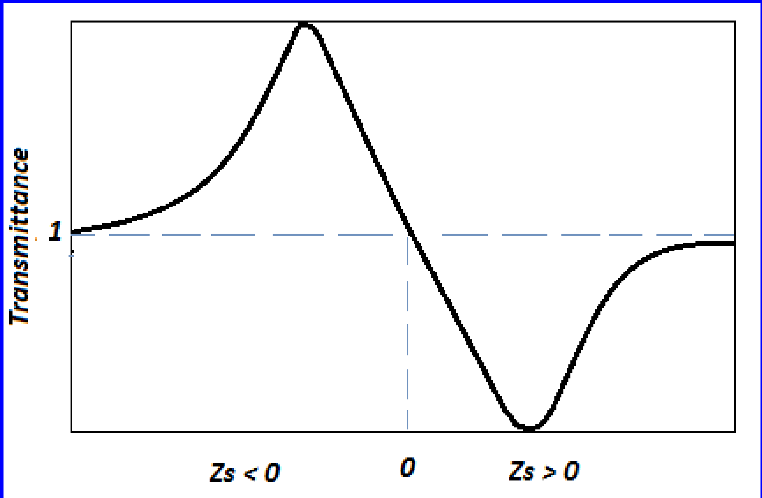


(a) (b)

Z-scan technique to measure nonlinear refractive index



$$T(L_a, \Delta\phi) \equiv \frac{I(z_s + L_a, \rho = 0, \Delta\phi)}{I(z_s + L_a, \rho = 0, \Delta\phi = 0)}$$



$$T_{\max} - T_{\min} \approx 0.406\,\Delta\phi$$

$$\Delta\phi = k_0 \bar{n}_2 I_0 \Delta L$$

Polarization dynamics of third-order processes

Isotropic media with inversion symmetry

By symmetry:

$$\chi^{(3)}_{xxxx} = \chi^{(3)}_{yyyy} = \chi^{(3)}_{zzzz},$$

$$\chi^{(3)}_{xxyy} = \chi^{(3)}_{xxzz} = \chi^{(3)}_{yyxx} = \chi^{(3)}_{yyzz} = \chi^{(3)}_{zzyy} = \chi^{(3)}_{zzxx},$$
$$\chi^{(3)}_{xyxy} = \chi^{(3)}_{xzxz} = \chi^{(3)}_{yzyz} = \chi^{(3)}_{zxzx} = \chi^{(3)}_{zyzy} = \chi^{(3)}_{yxyx},$$
$$\chi^{(3)}_{xyyx} = \chi^{(3)}_{yxxy} = \chi^{(3)}_{xzzx} = \chi^{(3)}_{zxxz} = \chi^{(3)}_{yzzy} = \chi^{(3)}_{zyyz}.$$

Rotational invariance:

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

etc. General form:

$$\begin{aligned} \chi^{(3)}_{ijkl} &= \chi^{(3)}_{xxyy}\delta_{ij}\delta_{kl} \\ &\quad + \chi^{(3)}_{xyxy}\delta_{ik}\delta_{jl} + \chi^{(3)}_{xyyx}\delta_{il}\delta_{jk} \end{aligned} \tag{5}$$

In particular, for self-focusing

$$\begin{aligned} \chi^{(3)}_{ijkl} &= \chi^{(3)}_{xxyy}(\delta_{ij}\delta_{kl} + \delta_{il}\delta_{jk}) \\ &\quad + \chi^{(3)}_{xyxy}\delta_{ik}\delta_{jl}. \end{aligned} \tag{6}$$

Nonlinear polarization in Maker& Terhune notations:

$$\mathscr{P}_{NL} = \varepsilon_0[A(\mathscr{E}\cdot\mathscr{E}^*)\mathscr{E} + \tfrac{1}{2}B(\mathscr{E}\cdot\mathscr{E})\mathscr{E}^*]$$

where two independent coefficients are introduced:

$$A \equiv \frac{3}{2}\chi^{(3)}_{xxyy}, \qquad B \equiv \frac{3}{2}\chi^{(3)}_{xyxy}$$

Evolution of elliptical polarization in isotropic nonlinear media

$$\mathscr{E} = \mathscr{E}_{+}\mathbf{e}_{+} + \mathscr{E}_{-}\mathbf{e}_{-}$$

Nonlinear polarization field in circular basis

$$\mathscr{P}_{NL} = \mathscr{P}_{NL}^{(+)}\mathbf{e}_{+} + \mathscr{P}_{NL}^{(-)}\mathbf{e}_{-}$$

$$\mathscr{P}_{NL}^{(\pm)} = \varepsilon_0 [\underbrace{A|\mathscr{E}_{\pm}|^2}_{\text{SPM}} + \underbrace{(A+B)|\mathscr{E}_{\mp}|^2}_{\text{CPM}}]\mathscr{E}_{\pm}$$

- SPM $\Longleftrightarrow$ self-phase modulation;
- CPM $\Longleftrightarrow$ cross-phase modulation.

Nonlinear susceptibility:

$$\chi_{NL}^{(\pm)} = A|\mathscr{E}_\pm|^2 + (A+B)|\mathscr{E}_\mp|^2$$

Effective refractive index:

$$n_\pm^2 = 1 + \chi_L + \chi_{NL}^{(\pm)}$$

Plane-wave propagation:

$$\frac{\partial^2 E_\pm}{\partial t^2} - \frac{n_\pm^2}{c^2}\frac{\partial^2 E_\pm}{\partial z^2} = 0,$$

- Each circular polarization propagates unchanged
- Each circular polarization has a slightly different refractive index

Elliptic polarization evolves as:

$$\mathbf{E}(z,t) = [\mathscr{E}_+ e^{i\Delta n \omega z/2c}\mathbf{e}_+ + \mathscr{E}_- e^{-i\Delta n \omega z/2c}\mathbf{e}_-]e^{i\omega(\bar{n}z/c-t)}$$

average refractive index

$$\bar{n} = n_L + \frac{(2A+B)}{4n_L}(|\mathscr{E}_+|^2 + |\mathscr{E}_-|^2),$$

nonlinear birefringence

$$\Delta n = n_+ - n_- = \frac{B}{2n_L}(|\mathscr{E}_-|^2 - |\mathscr{E}_+|^2),$$

Polarization rotation:

$$\mathbf{E}(z,t) = [\mathcal{E}_+\mathbf{e}_+(z) + \mathcal{E}_-\mathbf{e}_-(z)]e^{i\omega(\bar{n}z/c-t)}$$

with

$$\mathbf{e}_\pm(z) = \frac{\mathbf{e}_x(z) \pm i\mathbf{e}_y(z)}{\sqrt{2}}$$

where

$$\mathbf{e}_x(z) = \cos(\Delta n\omega z/2c)\mathbf{e}_x + \sin(\Delta n\omega z/2c)\mathbf{e}_y$$

$$\mathbf{e}_y(z) = \cos(\Delta n\omega z/2c)\mathbf{e}_y - \sin(\Delta n\omega z/2c)\mathbf{e}_x$$

Electro-optical Kerr effect

- Mixing of two optical and two dc fields:

$$\mathscr{P}_{NL} = 3\varepsilon_0[\chi^{(3)}_{xxyy}\mathscr{E}(\mathbf{E}_0\cdot\mathbf{E}_0) + 2\chi^{(3)}_{xyxy}\mathbf{E}_0(\mathscr{E}\cdot\mathbf{E}_0)]$$

Assuming

- $\mathbf{E}_0 = E_0\mathbf{e}_x$;
- $\mathscr{E} = \mathscr{E}_x\mathbf{e}_x + \mathscr{E}_y\mathbf{e}_y$

$$\mathscr{P}_{NLx} = 3\varepsilon_0\chi^{(3)}_{xxxx}E_0^2\mathscr{E}_x$$
$$\mathscr{P}_{NLy} = 3\varepsilon_0\chi^{(3)}_{xxyy}E_0^2\mathscr{E}_y$$

Plane-wave propagation:

$$\frac{\partial^2 E_{x,y}}{\partial t^2} - \frac{n_{x,y}^2}{c^2}\frac{\partial^2 E_{x,y}}{\partial z^2} = 0$$

Plane-wave solutions:

$$E_{x,y}(z,t) = \mathscr{E}_{x,y} e^{i(k_{x,y}z - \omega t)},$$

Wave numbers:

$$k_{x,y} = \frac{n_{x,y}\omega}{c}.$$

Assume the same linear refractive index n for both polarizations

Evolution of elliptic polarization:

$$\mathbf{E}(z,t) = \mathscr{E}_x[\mathbf{e}_x + \mathbf{e}_y \tan\theta e^{-i\Delta n\omega z/c}]e^{i\omega(n_x z/c - t)}$$

where

- $\tan\theta = \mathscr{E}_y/\mathscr{E}_x$
- $\Delta n = 3\chi^{(3)}_{xyxy}E_0^2/n$
- Polarization rotation through angle:

$$\Delta\phi_L = \frac{\Delta n\omega L}{c} = \frac{2\pi K E_0^2}{n}L$$

- Kerr constant:

$$K = \frac{\Delta n}{\lambda E_0^2}$$