

NONLINEAR OPTICS

Ch. 5 3rd ORDER NONLINEARITIES

- I. **Optical Kerr Effect** : self-focusing, nonlinear Schrödinger equation, self-phase modulation, solitons
- II. **Four-wave Mixing**
- III. **Raman Scattering** : spontaneous and stimulated Raman scattering, Raman amplification, Raman laser
- IV. **Brillouin Scattering** : spontaneous and stimulated Brillouin scattering

NONLINEAR OPTICS

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- I. **Optical Kerr Effect** : self-focusing, nonlinear Schrödinger equation, self-phase modulation, solitons

Energy transfer between a wave and the medium

Time-averaged power density transferred from the EM wave to the medium : $-\frac{\partial W}{\partial t} = \langle \vec{E} \cdot \frac{\partial \vec{P}}{\partial t} \rangle$

$$\begin{aligned} \vec{E}(t) &= \vec{E}(\omega)e^{-i\omega t} + CC \\ \vec{P}(t) &= \vec{P}(\omega)e^{-i\omega t} + CC \end{aligned} \quad \left/ \quad \begin{aligned} \langle \vec{E} \cdot \frac{\partial \vec{P}}{\partial t} \rangle &= -i\omega [\vec{E}(-\omega) \cdot \vec{P}(\omega) - \vec{E}(\omega) \cdot \vec{P}(-\omega)] \\ \vec{P}(\omega) &= \epsilon_0 \underline{\underline{\chi^{(3)}}}(\omega_P, -\omega_P, \omega) \vec{e}_P \vec{e}_P \vec{e} |E(\omega_P)|^2 E(\omega) \end{aligned}$$

$$\langle \vec{E} \cdot \frac{\partial \vec{P}}{\partial t} \rangle = -i\omega \epsilon_0 \left[\vec{e} \cdot \underline{\underline{\chi^{(3)}}}(\omega_P, -\omega_P, \omega) \vec{e}_P \vec{e}_P \vec{e} - \vec{e} \cdot \underline{\underline{\chi^{(3)*}}}(\omega_P, -\omega_P, \omega) \vec{e}_P \vec{e}_P \vec{e} \right] |E(\omega_P)|^2 |E(\omega)|^2$$

$$\chi^{(3)} = \chi^{(3)'} + i \chi^{(3)''}$$

Real part Imaginary part

$$\langle \vec{E} \cdot \frac{\partial \vec{P}}{\partial t} \rangle = 2\omega \epsilon_0 \vec{e} \cdot \chi^{(3)''}(\omega_P, -\omega_P, \omega) \vec{e}_P \vec{e}_P \vec{e} |E(\omega_P)|^2 |E(\omega)|^2$$

Conclusion :
If $\chi^{(3)}$ is purely real =
NO energy transfer
between the wave
and the material

I- Optical Kerr Effect

- Nonlinear refractive index**

$$\begin{aligned} \vec{E}(t) &= \vec{E}(\omega)e^{-i\omega t} + CC & \vec{P}_L(\omega) &= \epsilon_0 \underline{\underline{\chi^{(1)}}}(\omega) \vec{e} E(\omega) \\ \vec{P}(t) &= \vec{P}(\omega)e^{-i\omega t} + CC & \vec{P}_{NL}(\omega) &= 3\epsilon_0 \underline{\underline{\chi^{(3)}}}(\omega, -\omega, \omega) \vec{e} \vec{e} \vec{e} |E(\omega)|^2 E(\omega) \end{aligned}$$

Linear + Nonlinear polarization:

$$\vec{P}(\omega) = \epsilon_0 \left[\underline{\underline{\chi^{(1)}}}(\omega) \vec{e} + 3 \underline{\underline{\chi^{(3)}}}(\omega, -\omega, \omega) \vec{e} \vec{e} \vec{e} |E(\omega)|^2 \right] E(\omega)$$

$$\vec{P}(\omega) = \epsilon_0 \left[\chi_{eff}^{(1)} + \chi_{eff}^{(3)} |A(\omega)|^2 \right] A(\omega) e^{ikz}$$

Real part : $\chi_{eff}^{(3)'}$ induces a modification of the refractive index

$$n^2(\omega) = 1 + \underbrace{\chi_{eff}^{(1)} + \chi_{eff}^{(3)' |A(\omega)|^2}}_{n_0^2(\omega)}$$

Imaginary part : $\chi_{eff}^{(3)''}$

induces a modification of the absorption

$$n(\omega) = n_0(\omega) \sqrt{1 + \underbrace{\frac{\chi_{eff}^{(3)'}}{2n_0^3 \epsilon_0 c} I(\omega)}_{\epsilon \ll 1}} \approx n_0(\omega) + n_2 I(\omega)$$

I- Optical Kerr Effect

- Nonlinear refractive index**

$$n \approx n_0 + n_2 I$$

$$n_2 = \frac{\chi_{eff}^{(3)}}{4n_0^2 \epsilon_0 c}$$

Example :

Pure Silica $n_0=1,45$
and $n_2= 3 \cdot 10^{-20} \text{ m}^2/\text{W}$

- Nonlinear Phase**

Temporal propagation

$$\phi_{NL}(t) = 2\pi \frac{n_2 I(t) L}{\lambda}$$

$$\Delta\omega = -\frac{d\phi_{NL}}{dt} = -2\pi \frac{n_2 L}{\lambda} \frac{dI(t)}{dt}$$

Consequences : time-varying phase, Self-phase modulation, temporal soliton

Spatial propagation

$$\phi_{NL}(\vec{r}) = 2\pi \frac{n_2 I(\vec{r}) L}{\lambda}$$

Consequences : Self-focusing or self-defocusing effect, filamentation, spatial soliton

I- Optical Kerr Effect

- Self-Phase Modulation (SPM) effect**

Nonlinear wave equation, assuming an instantaneous response of the material and neglecting linear distortion effects (dispersion and diffraction)

$$\frac{\partial A(\rho, z)}{\partial z} = ik_0 n_2 I(\rho, z) A(\rho, z) \quad \begin{cases} \rho = t & \text{Time} \\ \text{or} \\ \rho = \mathbf{r} & \text{Space} \end{cases}$$

Lossless medium $\rightarrow I(\rho, z) = \text{Cste at a fixed } t \text{ or } \mathbf{r}$

Solution $\rightarrow A(\rho, z) = A(\rho, 0) e^{ik_0 n_2 I(\rho) z}$
 $= A(\rho, 0) e^{i\Phi_{NL}(\rho, z)}$

$\left[\begin{array}{l} |A(t, z)|^2 = |A(t, 0)|^2 \\ \text{or} \\ |A(\mathbf{r}, z)|^2 = |A(\mathbf{r}, 0)|^2 \end{array} \right.$

\Rightarrow **The phase matching is automatically fulfilled**

\Rightarrow **Assuming n_2 is a purely real quantity, the SPM induces a phase variation proportionally to the intensity**

\Rightarrow **The beam shape (in time or space) in intensity is conserved along the beam propagation (neglecting linear distortion effects)**

I- Optical Kerr Effect

• Self-Phase Modulation (SPM) effect

Nonlinear wave equation, assuming an instantaneous response of the material and neglecting linear distortion effects (dispersion and diffraction)

$$\frac{\partial A(\rho, z)}{\partial z} = ik_0 n_2 I(\rho, z) A(\rho, z) \quad \begin{cases} \rho = t & \text{Time} \\ \text{or} \\ \rho = \mathbf{r} & \text{Space} \end{cases}$$

Lossless medium $\rightarrow I(\rho, z) = \text{Cste at a fixed } t \text{ or } \mathbf{r}$

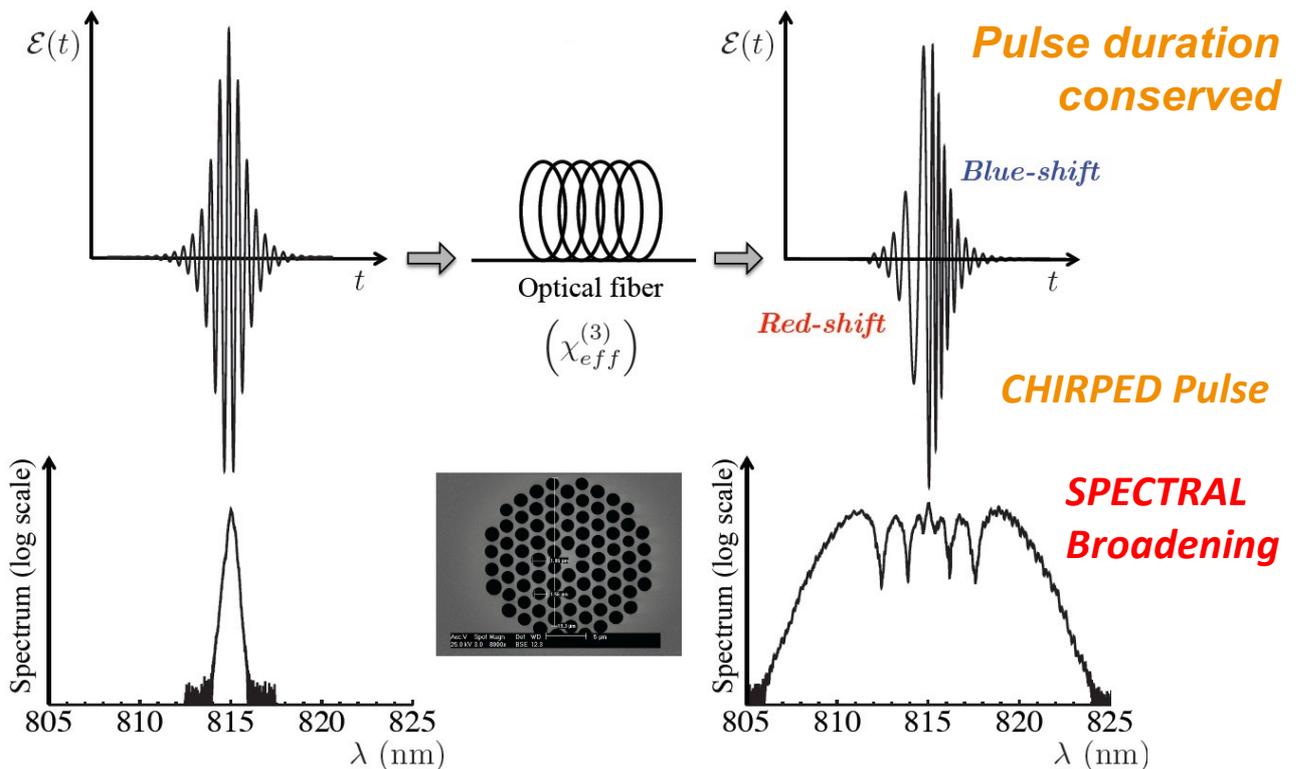
Solution $\rightarrow A(\rho, z) = A(\rho, 0)e^{ik_0 n_2 I(\rho)z}$
 $= A(\rho, 0)e^{i\Phi_{\text{NL}}(\rho, z)}$, $\rightarrow \begin{cases} |A(t, z)|^2 = |A(t, 0)|^2 \\ \text{or} \\ |A(\mathbf{r}, z)|^2 = |A(\mathbf{r}, 0)|^2 \end{cases}$

- Nonlinear phase of a transmitted pulse or beam $\Phi_{\text{NL}}(\rho, z) = k_0 n_2 I(\rho)z$
- Instantaneous frequency (case of a PULSE)

Spectral broadening + Frequency CHIRP of the pulse

$$\Delta\omega_{\text{NL}} \simeq -\frac{d\Phi_{\text{NL}}(t)}{dt} = -k_0 n_2 \frac{dI(t)}{dt} z.$$

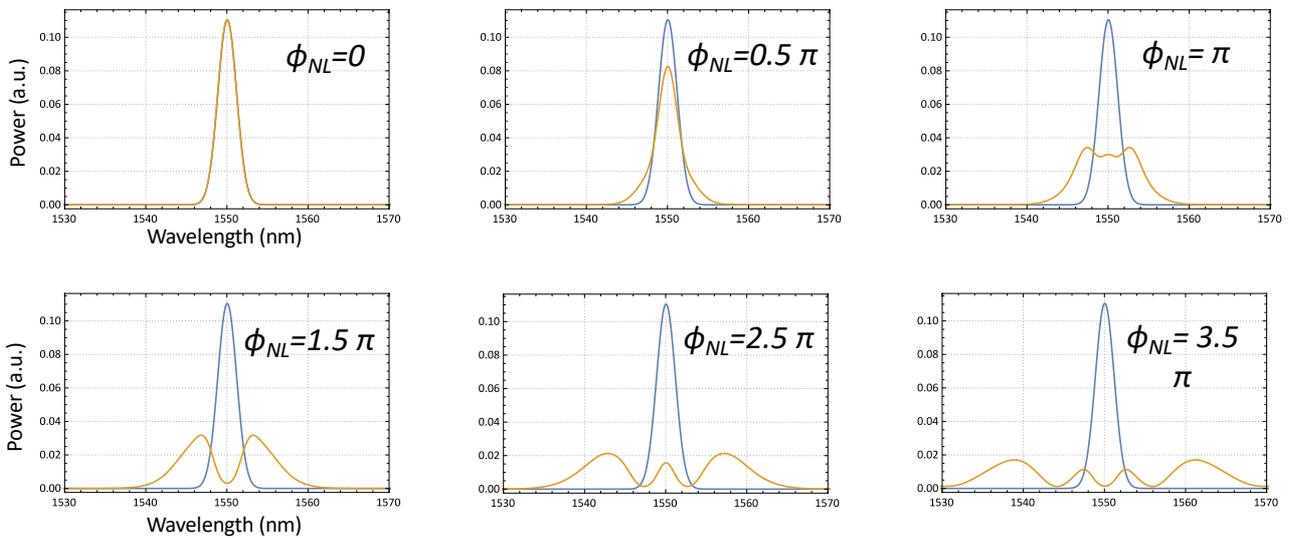
Self-Phase Modulation



From Margaux Barbier, LCF Manolia, PhD manuscript (2014)

Self-Phase Modulation

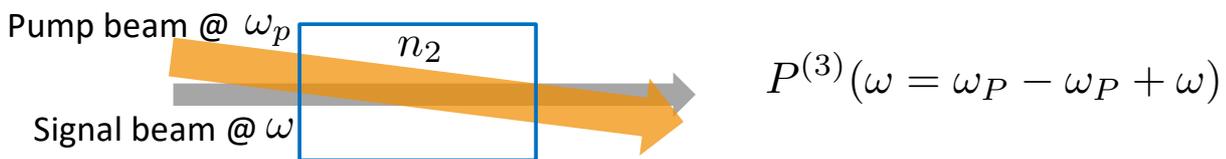
- Spectral broadening through SPM – Gaussian Pulse**



I- Optical Kerr Effect

- Cross-Phase Modulation (XPM) effect**

The optical Kerr effect is then driven by the intensity of a pump wave @ ω_P , leading to the modification of the phase experienced by a signal wave @ ω



$$P^{(3)}(\omega) = 6\epsilon_0\chi_{eff}^{(3)}|A_P(\rho, z)|^2 A(\rho, z) \quad \begin{cases} \rho = t & \text{Time} \\ & \text{or} \\ \rho = r & \text{Space} \end{cases}$$

Nonlinear phase for the signal

beam (pulse or beam) $\phi_{NL}(\rho, z) = 2k_0n_2I_P(\rho)z$

\Rightarrow **In case of XPM, the nonlinear phase shift is x 2 compared to SPM (consequence of the degeneracy factor)**

I- Optical Kerr Effect

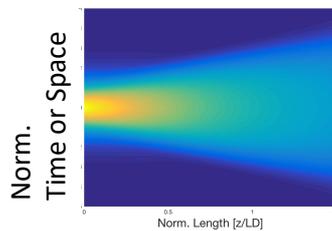
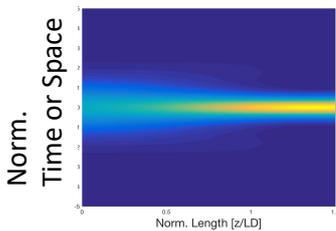
What have we learned ?

- propagation of a wave packet through a pure Kerr medium leads to a phase modification proportionally to the wave intensity.
- The phase matching condition is automatically fulfilled
- A pure Kerr and lossless medium is equivalent to a spatial or/and a temporal phase modulator (phase \propto Intensity) and leads to a spectral broadening

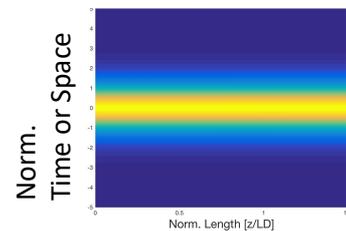
→ **SPATIAL / TEMPORAL KERR LENS EFFECT**

Question : interplays between linear and nonlinear Kerr effects?

Self-focusing & de-focusing effects



Soliton effect



→ **Nonlinear Schrödinger Equation**

I- Optical Kerr Effect

• Nonlinear Schrödinger Equation – Time domain

Propagation of a pulse : $\mathcal{E}(z, t) = A(z, t)e^{i\beta_0 z} e^{-i\omega_0 t} e + CC,$
 $\mathcal{P}_{NL}(z, t) = \mathcal{P}_{NL}(z, t)e^{i\beta_p z} e^{-i\omega_0 t} + CC.$

- We start with the wave equation in the frequency domain :

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r}, \omega) = \frac{\omega^2}{c^2} \underline{\underline{\epsilon}}(\mathbf{r}, \omega) \mathbf{E}(\mathbf{r}, \omega) + \omega^2 \mu_0 \mathbf{P}^{(NL)}(\mathbf{r}, \omega)$$



writing $\mathbf{E}(z, \omega) = e \tilde{A}(z, \omega - \omega_0) e^{i\beta_0 z}$
 $\mathbf{P}_{NL}(z, \omega) = \tilde{\mathcal{P}}_{NL}(z, \omega - \omega_0) e^{i\beta_p z}$

$$\frac{\partial^2 \tilde{A}(z, \omega - \omega_0)}{\partial z^2} + 2i\beta_0 \frac{\partial \tilde{A}(z, \omega - \omega_0)}{\partial z} + [\beta^2(\omega) - \beta_0^2] \tilde{A}(z, \omega - \omega_0) = -\omega^2 \mu_0 e \cdot \tilde{\mathcal{P}}_{NL}(z, \omega - \omega_0) e^{i\Delta\beta z}$$

I- Optical Kerr Effect

• Nonlinear Schrödinger Equation – Time domain

- Power series expansion of $\beta(\omega)^{\beta_0}$ $\beta(\omega) = \beta_0 + \beta_1 \Delta\omega + \frac{\beta_2}{2} \Delta\omega^2 + \dots$,
 $\Delta\omega \ll \omega_0$ $\beta^2(\omega) \simeq \beta_0^2 + 2\beta_0\beta_1 \Delta\omega + (\beta_1^2 + \beta_0\beta_2) \Delta\omega^2 + \dots$

- after substitution and following a Fourier transformation, the wave equation yields

$$\mathcal{E}(t) = \int \mathbf{E}(\omega) e^{-i\omega t} d\omega$$

$$\frac{\partial^2 A(z, t)}{\partial z^2} + 2i\beta_0 \frac{\partial A(z, t)}{\partial z} + 2i\beta_0\beta_1 \frac{\partial A(z, t)}{\partial t} - [\beta_1^2 + \beta_0\beta_2] \frac{\partial^2 A(z, t)}{\partial t^2} = \mu_0 e \cdot \left[-\omega_0^2 \Pi_{NL}(z, t) - 2i\omega_0 \frac{\partial \Pi_{NL}(z, t)}{\partial t} + \frac{\partial^2 \Pi_{NL}(z, t)}{\partial t^2} \right] e^{i\Delta\beta z}$$

- Time coordinate transformation

(introduction of a retarded time) : $\tau = t - z/v_g$.

$$v_g = 1/\beta_1$$

(Group velocity)

$$\frac{\partial^2 A(z, \tau)}{\partial z^2} - 2\beta_1 \frac{\partial^2 A(z, \tau)}{\partial z \partial \tau} + 2i\beta_0 \frac{\partial A(z, \tau)}{\partial z} - \beta_0\beta_2 \frac{\partial^2 A(z, \tau)}{\partial \tau^2} = \mu_0 e \cdot \left[-\omega_0^2 \Pi_{NL}(z, \tau) - 2i\omega_0 \frac{\partial \Pi_{NL}(z, \tau)}{\partial \tau} + \frac{\partial^2 \Pi_{NL}(z, \tau)}{\partial \tau^2} \right] e^{i\Delta\beta z}$$

I- Optical Kerr Effect

• Nonlinear Schrödinger Equation – Time domain

$$\frac{\partial^2 A(z, \tau)}{\partial z^2} - 2\beta_1 \frac{\partial^2 A(z, \tau)}{\partial z \partial \tau} + 2i\beta_0 \frac{\partial A(z, \tau)}{\partial z} - \beta_0\beta_2 \frac{\partial^2 A(z, \tau)}{\partial \tau^2} = \mu_0 e \cdot \left[-\omega_0^2 \Pi_{NL}(z, \tau) - 2i\omega_0 \frac{\partial \Pi_{NL}(z, \tau)}{\partial \tau} + \frac{\partial^2 \Pi_{NL}(z, \tau)}{\partial \tau^2} \right] e^{i\Delta\beta z}$$

- Narrow spectral linewidth : $\Delta\omega \ll \omega_0$

$$\left| \frac{\partial A(z, \tau)}{\partial \tau} \right| \ll \omega_0 A(z, \tau) \quad \& \quad \left| 2\beta_1 \frac{\partial^2 A(z, \tau)}{\partial z \partial \tau} \right| \ll 2 \left| \beta_0 \frac{\partial A(z, \tau)}{\partial z} \right|$$

- Slow variation of $P_{NL}(t)$

within a time period $T=2\pi/\omega_0$

$$\omega_0^2 \Pi_{NL}(z, \tau) \gg 2\omega_0 \frac{\partial \Pi_{NL}(z, \tau)}{\partial \tau} \gg \frac{\partial^2 \Pi_{NL}(z, \tau)}{\partial \tau^2}$$

- Slowly-varying amplitude approximation $\left| \frac{\partial^2 A(z, \tau)}{\partial z^2} \right| \ll \left| \beta_0 \frac{\partial A(z, \tau)}{\partial z} \right|$

$$\frac{\partial A(z, \tau)}{\partial z} + \frac{i\beta_2}{2} \frac{\partial^2 A(z, \tau)}{\partial \tau^2} = \frac{i\omega}{2\epsilon_0 n c} e \cdot \Pi_{NL}(z, \tau) e^{i\Delta k z}$$

I- Optical Kerr Effect

- **Nonlinear Schrödinger Equation – TEMPORAL domain**

In presence of Kerr effect

$$\Pi_{NL}(t, z) = \epsilon_0 \chi_{\text{eff}}^{(3)} |A(t, z)|^2 A(t, z) e$$

Assumption :
instantaneous
response of Kerr
effect $\gamma' = \frac{\omega_0}{2nc} \chi_{\text{eff}}^{(3)}$

NONLINEAR SCHRÖDINGER EQUATION

$$\frac{\partial A(\tau, z)}{\partial z} + \frac{i\beta_2}{2} \frac{\partial^2 A(\tau, z)}{\partial \tau^2} - i\gamma' |A(\tau, z)|^2 A(\tau, z) = 0$$

2rd order
dispersion
effect

Nonlinear Kerr term
(phase variation α
Intensity)

I- Optical Kerr Effect

- **Nonlinear Schrödinger Equation – SPATIAL domain**

In presence of Kerr effect

$$\Pi_{NL}(\mathbf{r}, z) = \epsilon_0 \chi_{\text{eff}}^{(3)} |A(\mathbf{r}, z)|^2 A(\mathbf{r}, z) e$$

$$\gamma' = \frac{\omega_0}{2nc} \chi_{\text{eff}}^{(3)}$$

NONLINEAR SCHRÖDINGER EQUATION

$$\frac{\partial A(\mathbf{r}, z)}{\partial z} + \frac{1}{2ik} \Delta_T A(\mathbf{r}, z) - i\gamma |A(\mathbf{r}, z)|^2 A(\mathbf{r}, z) = 0$$

Diffraction
effect

Nonlinear Kerr term
(phase variation α
Intensity)

I- Optical Kerr Effect

- Dispersion length & Nonlinear length**

Variables transformation $T = \frac{\tau}{\tau_0}$ $A(t, z) = \sqrt{\frac{I_0}{2nc\epsilon_0}} u(t, z)$

$$\frac{\partial u}{\partial z} + \frac{i \text{sign}(\beta_2)}{2L_D} \frac{\partial^2 u}{\partial T^2} - i \frac{|u|^2 u}{L_{NL}} = 0$$

with τ_0 : pulse duration
 P_0 : peak power of the pulse

$$L_D = \frac{\tau_0^2}{|\beta_2|} \text{ Dispersion length}$$

$$L_{NL} = \frac{1}{k_0 n_2 I_0} \text{ Nonlinear length}$$

Once the interaction length is set :

$L \ll L_D$ The dispersive effect can be neglected

$L \ll L_{NL}$ The optical Kerr effect can be neglected

I- Optical Kerr Effect

- Nonlinear Schrödinger Equation**

- NLSE : physics insights**

$$\frac{\partial u}{\partial z} + \frac{i \text{sign}(\beta_2)}{2L_D} \frac{\partial^2 u}{\partial T^2} - i \frac{|u|^2 u}{L_{NL}} = 0$$

Dispersion
effect

Kerr
LENS



$$\frac{\partial u}{\partial z} + \frac{i \text{sign}(\beta_2)}{2L_D} \frac{\partial^2 u}{\partial T^2} - i \frac{|u|^2 u}{L_{NL}} = 0$$

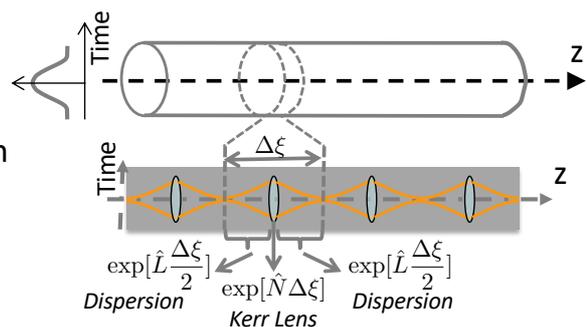
Kerr effect

$$u(T, z) = u(T, 0) e^{i \frac{|u(T)|^2}{L_{NL}} z}$$

APPLICATION :

$$\frac{\partial u}{\partial \xi} + \hat{L}u + \hat{N}u = 0$$

Numerical simulation
« Spilt Step Fourier
Method »



I- Optical Kerr Effect

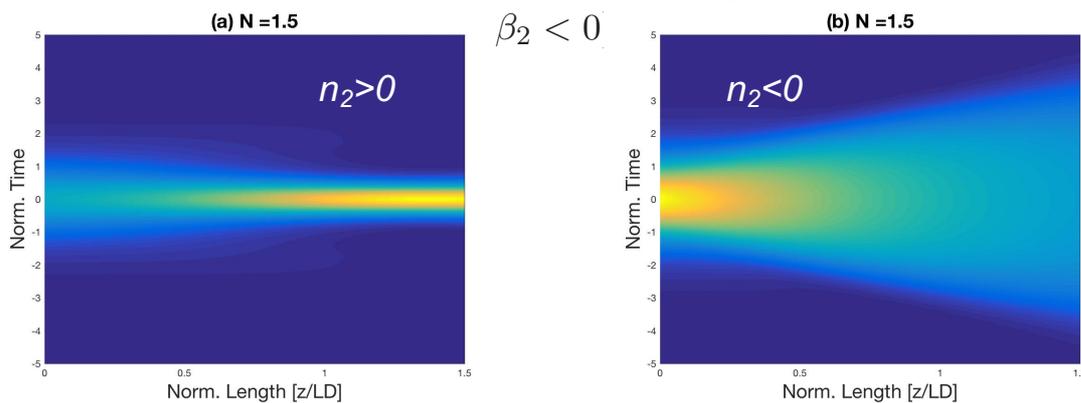
Nonlinear Schrödinger Equation

Variables transformation $\xi = z/L_D$ $N^2 = L_D/L_{NL}$

$$i \frac{\partial u}{\partial \xi} - \text{sign}(\beta_2) \frac{1}{2} \frac{\partial^2 u}{\partial T^2} + N^2 |u|^2 u = 0$$

- APPLICATIONS : Self-focusing & de-focusing effects**
Anomalous dispersion regime

$$N > 1$$



I- Optical Kerr Effect

Nonlinear Schrödinger Equation

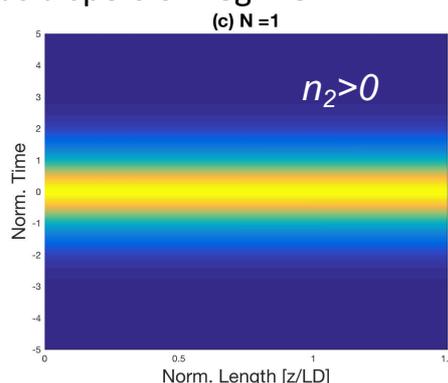
Variables transformation $\xi = z/L_D$ $N^2 = L_D/L_{NL}$

$$i \frac{\partial u}{\partial \xi} - \text{sign}(\beta_2) \frac{1}{2} \frac{\partial^2 u}{\partial T^2} + N^2 |u|^2 u = 0$$

- APPLICATIONS : Soliton effect**

Anomalous dispersion regime

$$\beta_2 < 0$$



$$N = 1$$

Strict compensation of the dispersive effect by the optical Kerr effect

⇒ Propagation of the pulse without deformation

⇒ Soliton solutions :

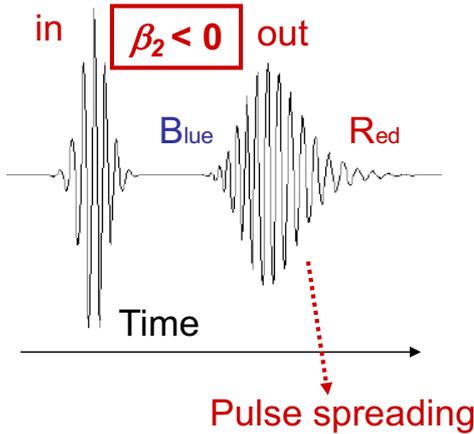
$$u(\rho) = N \text{sech}(\rho)$$

For further reading : Nonlinear Fiber Optics, Ch.5 by Govind P. Agrawal

I- Optical Kerr Effect

- **Pulse propagation : influence of the DISPERSION EFFECT and the OPTICAL KERR EFFECT n_2 ...**

Pulse propagation in a dispersive medium (anomalous dispersion, $D > 0$)



Pulse propagation in a NL medium ($n_2 > 0$)

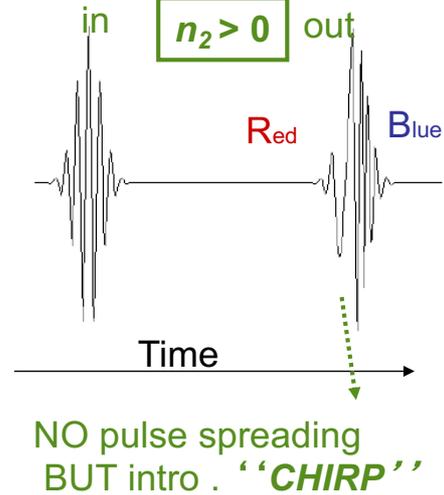


Schéma pulse copié de <http://www.sccs.swarthmore.edu/users/02/lisal/physics/presentations/soliton.pdf>

I- Optical Kerr Effect

- **Temporal Soliton**

Pulse propagation in a NL medium ($n_2 > 0$) and dispersive ($\beta_2 < 0$)

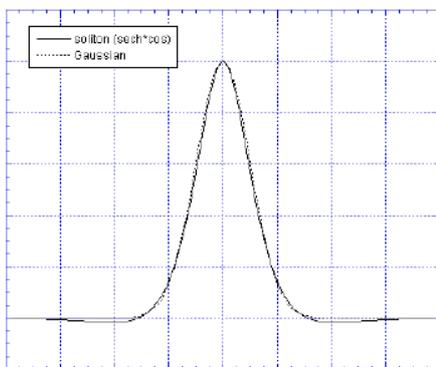
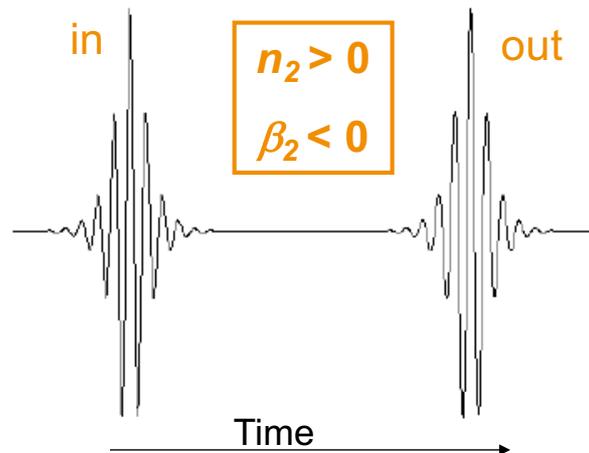


Figure 3: Envelope of soliton solution to NLSE compar envelope of a Gaussian pulse.

Soliton solution = « sech » pulse shape with no deformation



Full compensation between group-velocity dispersion and the effect of self-phase modulation

<http://www.sccs.swarthmore.edu/users/02/lisal/physics/presentations/soliton.pdf>

I- Optical Kerr Effect

- **NL Schrödinger Eq. :**

temporal Soliton

- **fundamental Soliton N=1 :**

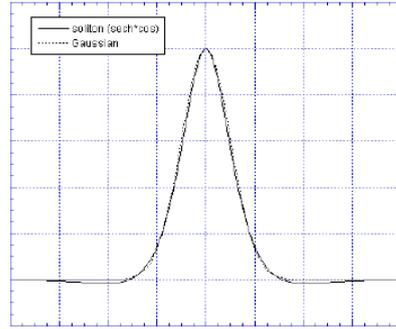
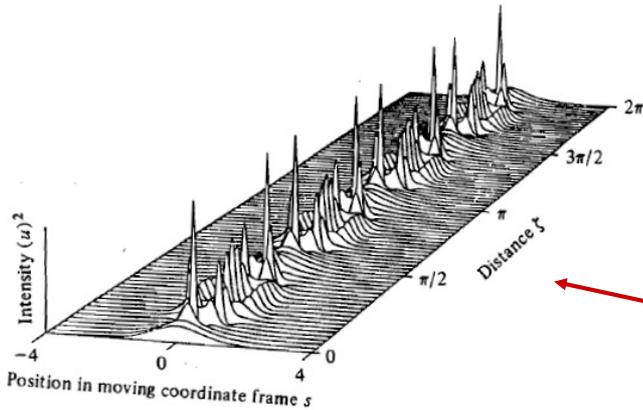


Figure 3: Envelope of soliton solution to NLSE compared with envelope of a Gaussian pulse.

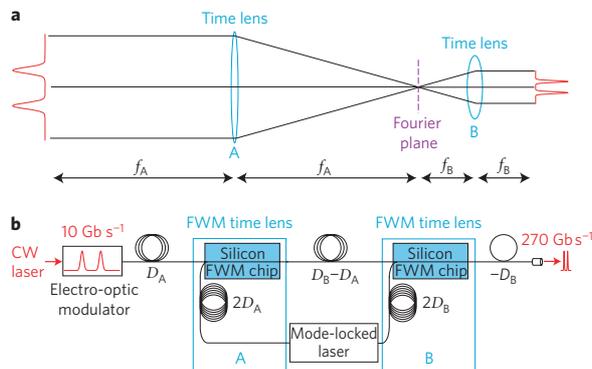


- **Soliton N=4 : shape**

Fig. 7.8 Computer-generated solutions of the nonlinear Schrödinger equation showing the evolution of an $N = 4$ soliton as it propagates over a distance equal to four soliton periods. (After Blow and Doran, 1987.)

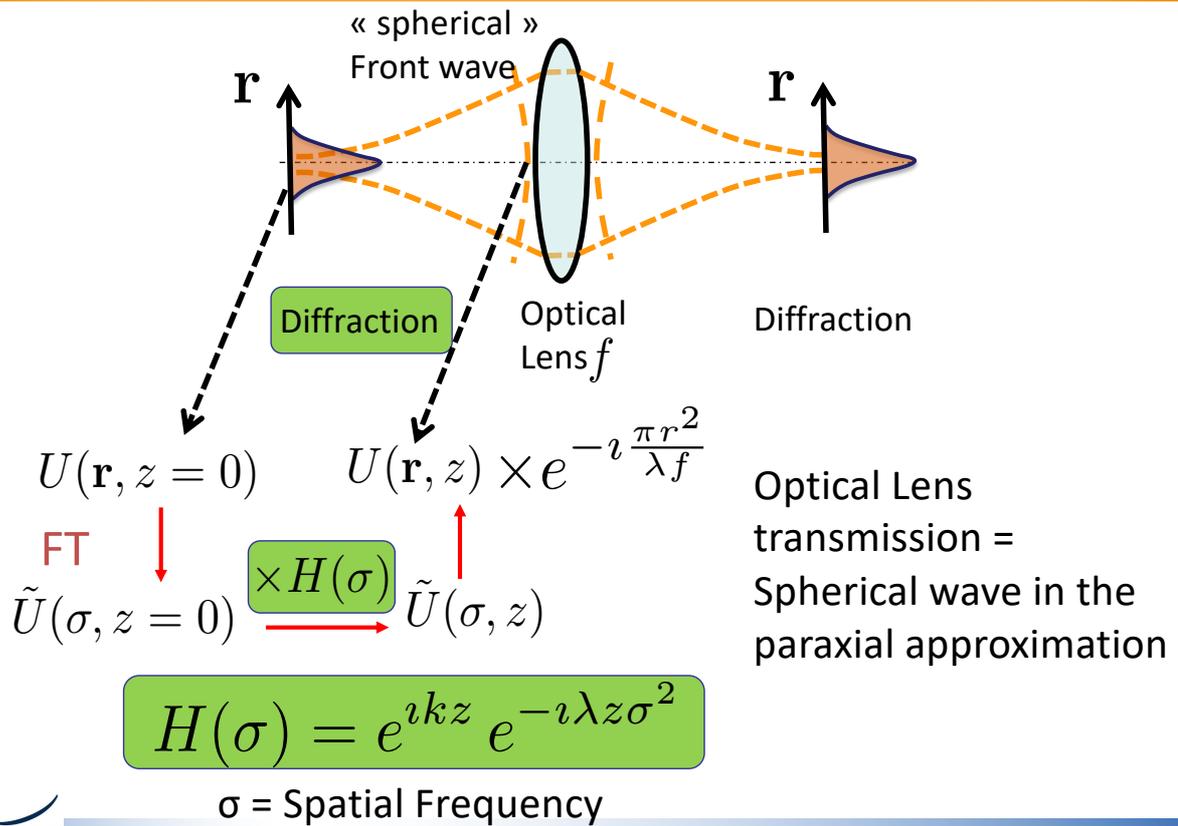
Fig. extraite du livre de Butcher & Cotter « The elements of nonlinear optics »

SUPPLEMENT : Temporal Imaging

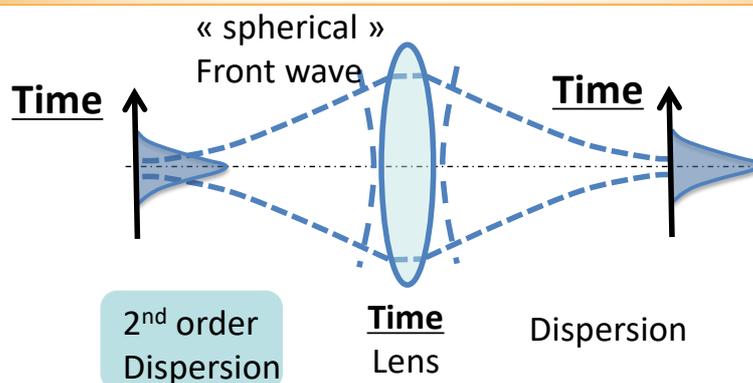


Forster et al., Nat Phot. (2009)

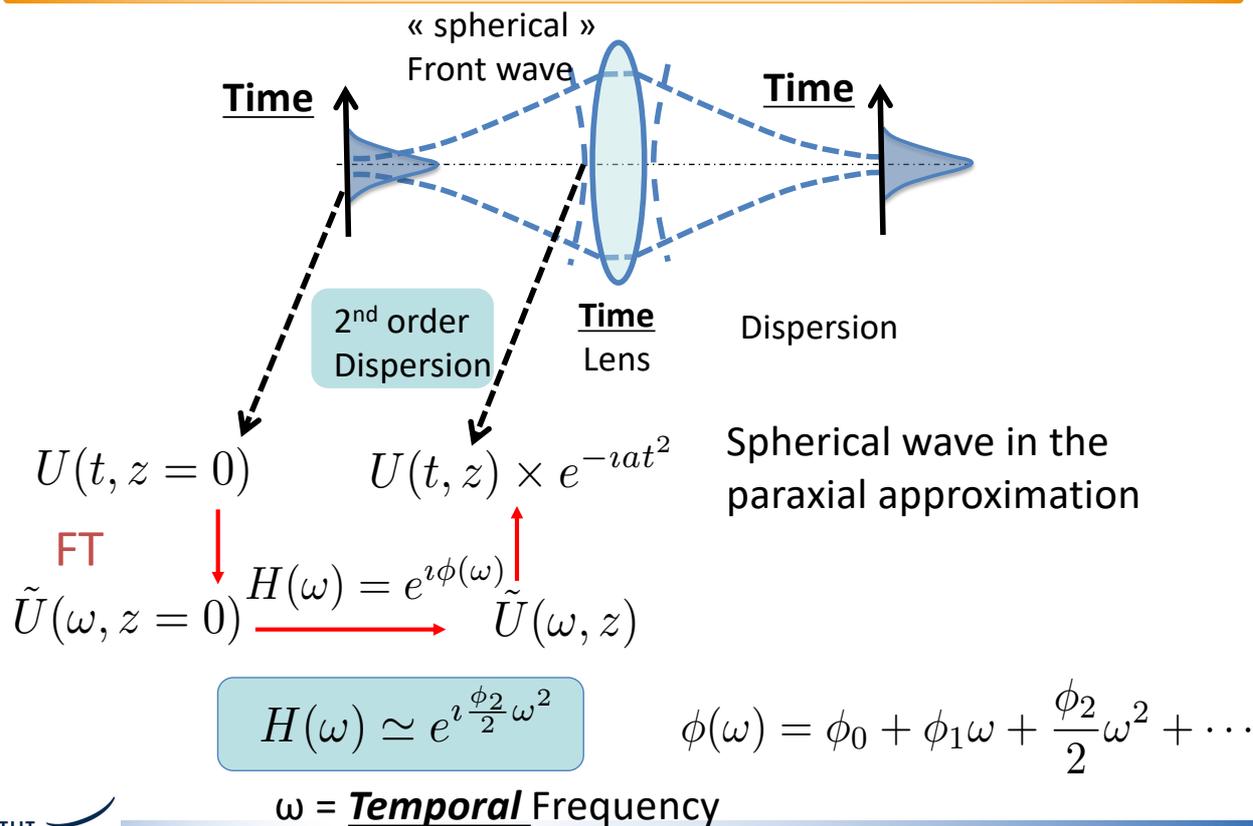
Spatial Imaging



Temporal Imaging



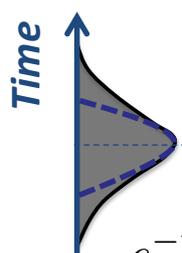
Temporal Imaging



Optical Kerr-Lens effect

Gaussian pulse shape

Optical Kerr Effect :

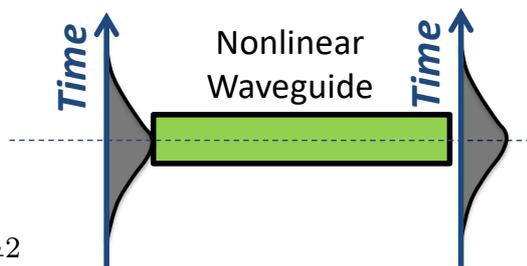


→ Paraxial approximation

$$e^{-t^2/T^2} \simeq 1 - t^2/T^2$$

$$\phi_{NL}(t) \simeq -\frac{2\pi}{\lambda} n_2 \frac{I_0}{\sqrt{1 + \left(\frac{\phi_2}{T_0^2}\right)^2}} \frac{t^2}{T^2} L$$

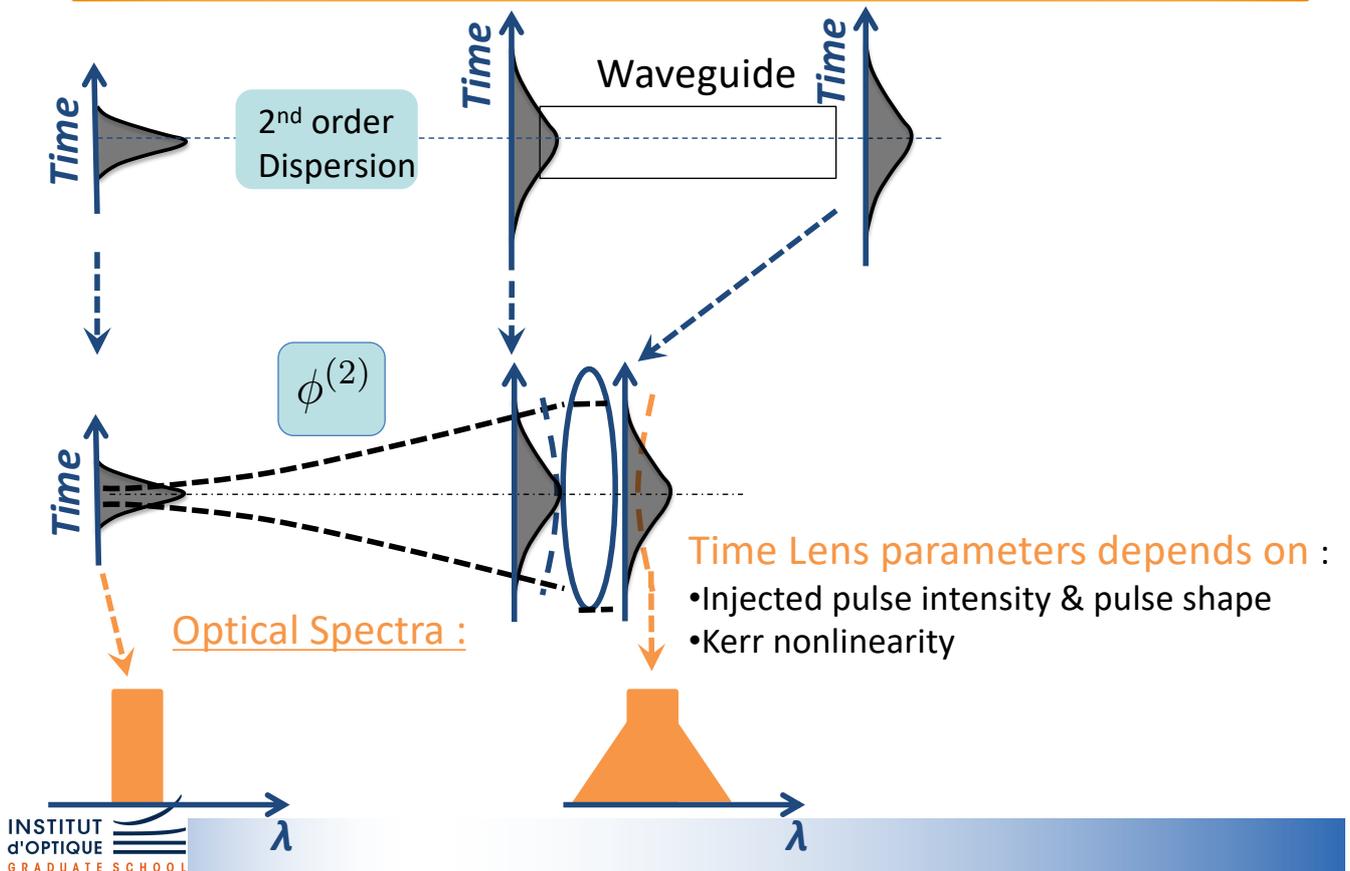
$$U(t, z') = U(t, z) \times e^{i\frac{2\pi}{\lambda} n_2 I(t) L}$$



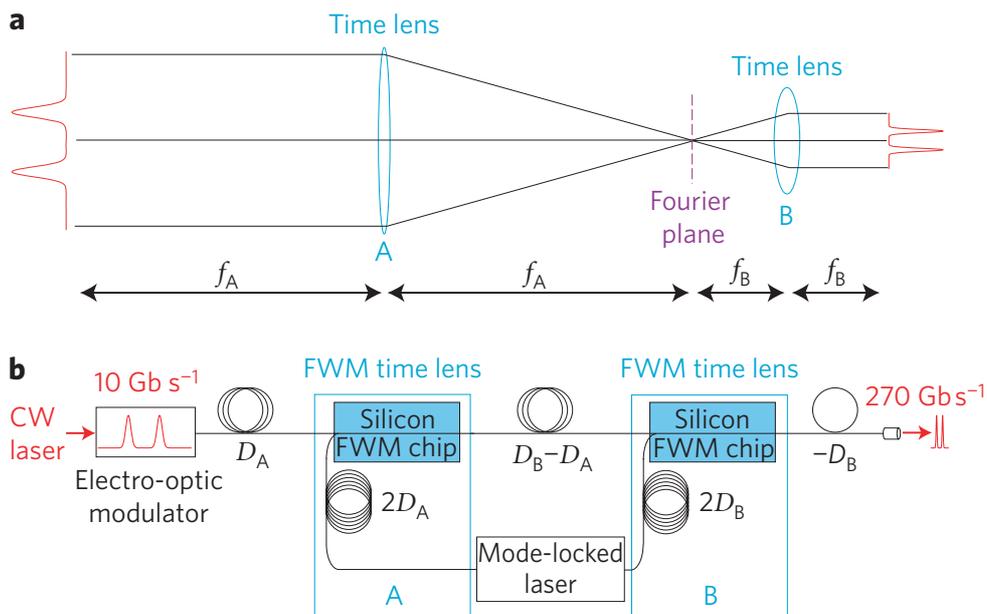
$$\rightarrow U(t, z) \times e^{-iat^2}$$

→ Optical Kerr effect = Temporal Lens

Optical Kerr-Lens : Gaussian beams



Application : time-domain telescope



Forster et al., Nat Phot. (2009)