Impulsions lumineuses ultracourtes et applications en diagnostic optique

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Outline

- Ultrashort pulses – some orders of magnitude
- Ultrashort pulse generation : Mode-locked oscillators
- Frequency comb spectroscopy
- Energy scaling
- Coherent Raman spectroscopy in reactive media
- Ultrafast imaging

Impulsions gaussienne : E(t) champ électrique

Domaine temporel :

$$E(t) = E_0 \exp\left(-\frac{t^2}{2T_0^2}\right)\exp(-i\omega_0 t)$$

$$I(t) = I_0 \exp\left(-\frac{t^2}{T_0^2}\right)$$

TF

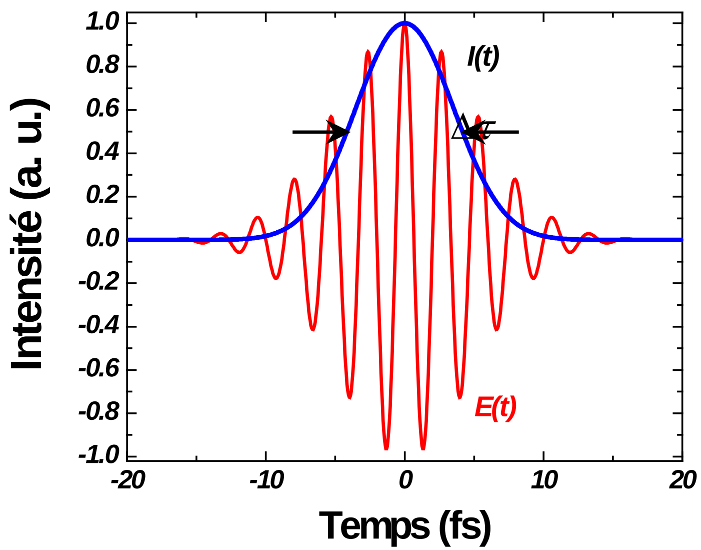
Domaine fréquentiel :

$$E(\omega) = E_0 T_0 \sqrt{2\pi}\exp\left(-\frac{(\omega-\omega_0)^2 T_0^2}{2}\right)$$

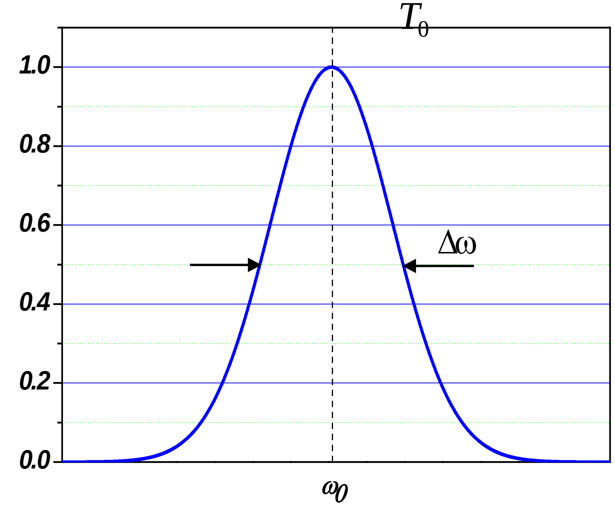
$$I(\omega) = 2\pi T_0^2 I_0 \exp\left(-(\omega-\omega_0)^2 T_0^2\right)$$

Largeurs à mi-hauteur : $\Delta\tau = 2\sqrt{\ln 2}\,T_0$

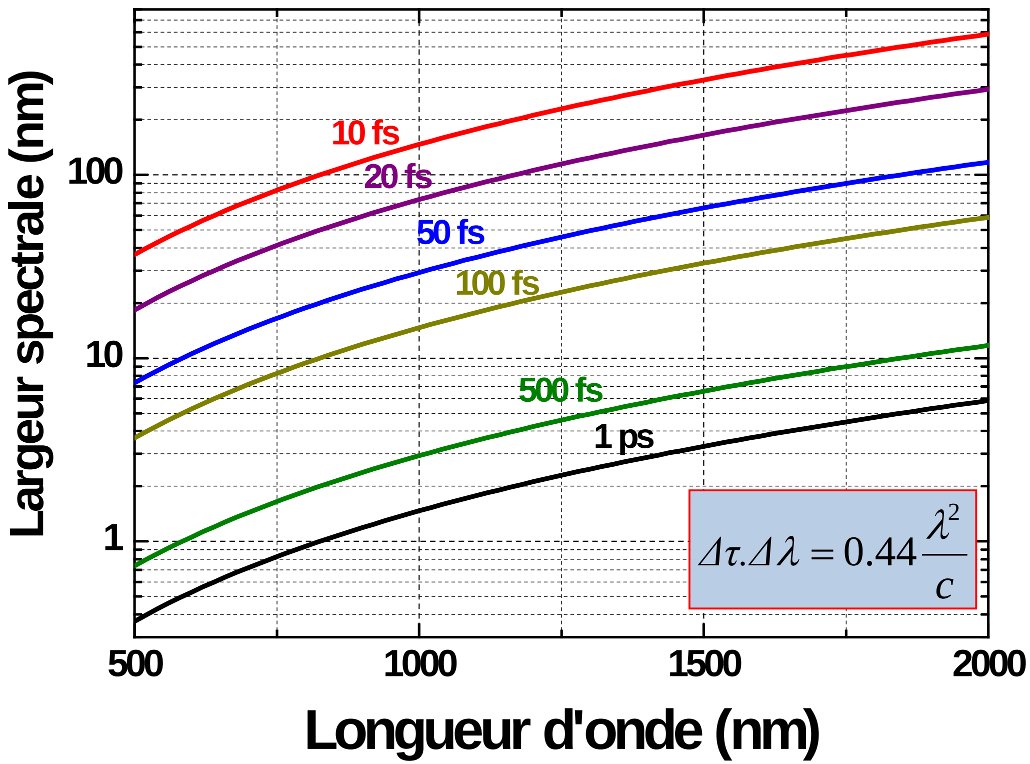
$$\Delta\omega = \frac{2\sqrt{\ln 2}}{T_0}$$



$$\Delta\tau.\Delta\nu = \frac{2\ln 2}{\pi} = 0.44$$



Ultrashort pulses ⇒ Large bandwidth



$\Delta\tau\ .\ \Delta\nu = K$

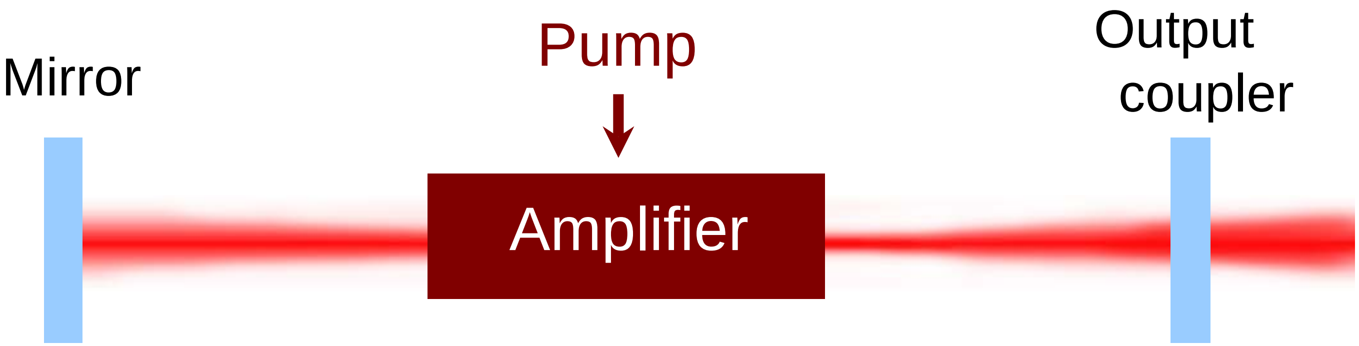
K = 0.44 : Gaussian pulse

K=0.315 : $sech^2$ pulse

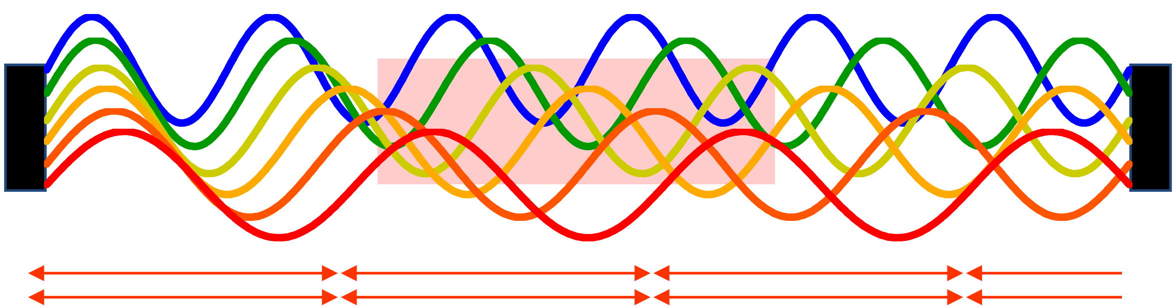
Gaussian pulse width 100 fs at 1 µm ⇒ spectral width 14.6 nm

Pulse width 20 fs ⇒ spectral width 73 nm

Ultrashort pulses generation – Mode-locking

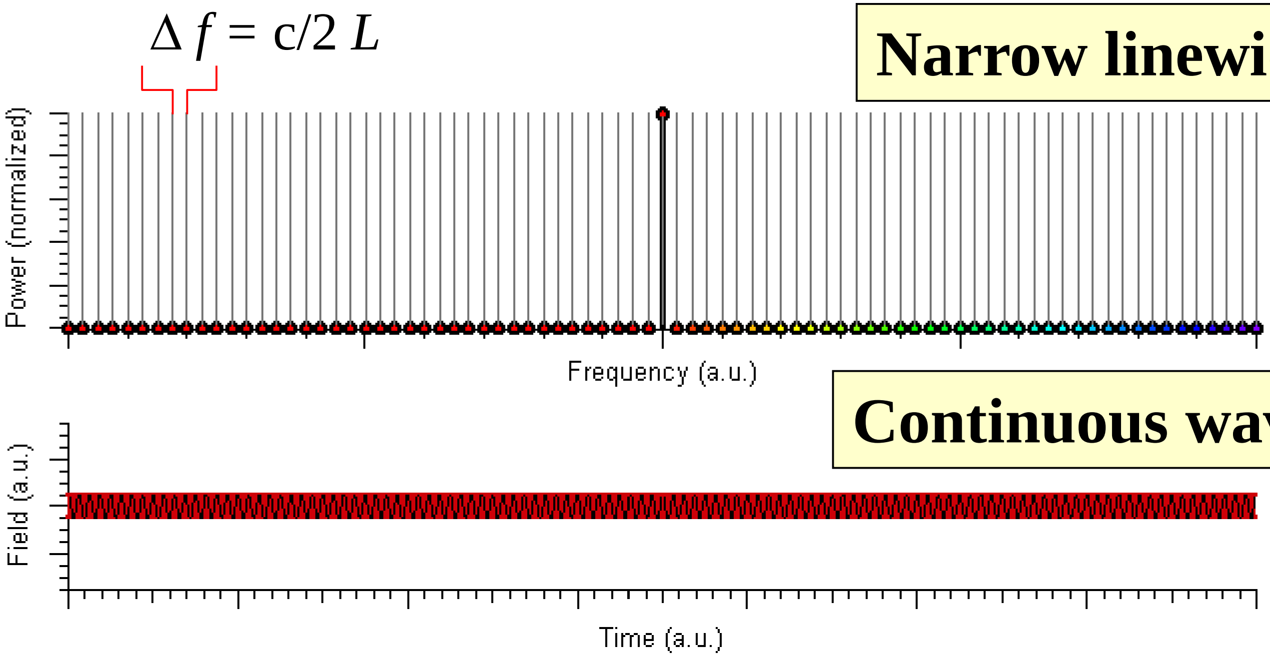


Longitudinal modes – spatial presentation



$\lambda_m = 2\,L/m$ $\Delta f = c/2\,L$

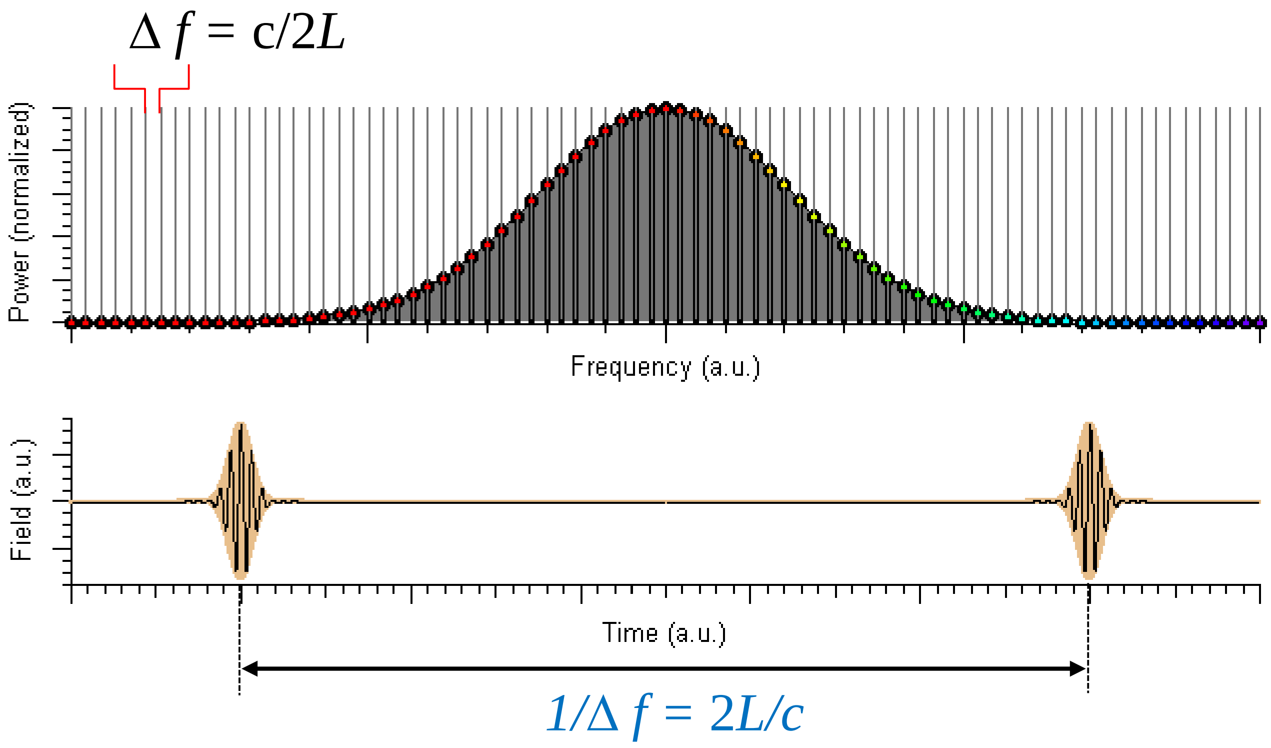
Time-bandwidth relation



Narrow linewidth

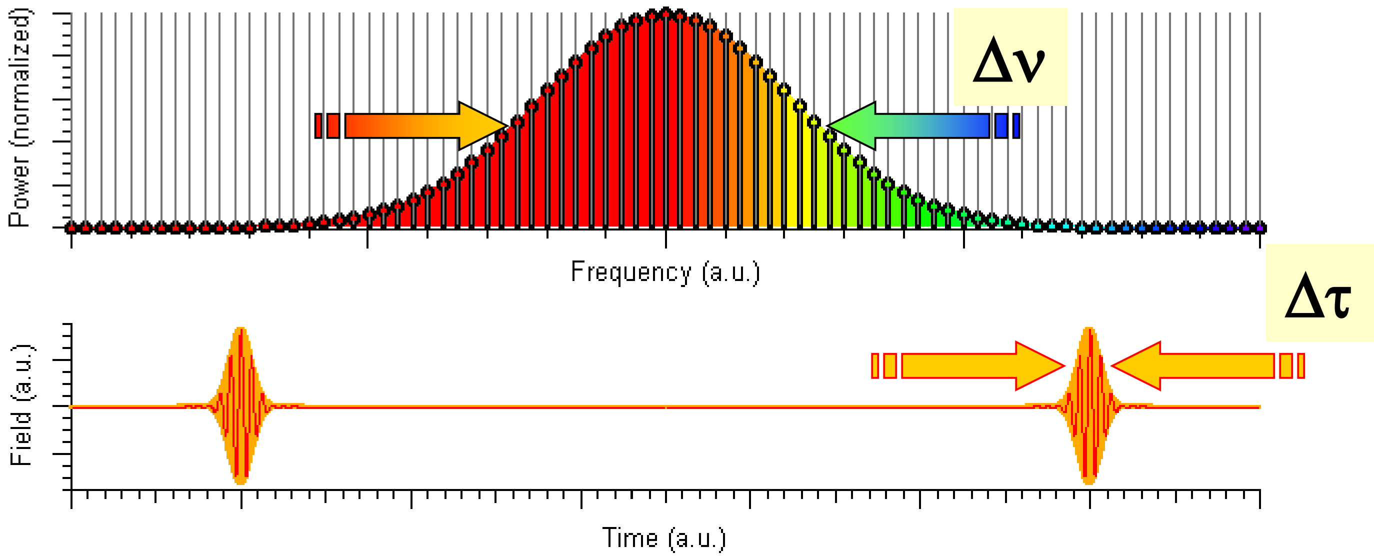
Continuous wave

Time-bandwidth relation



Need to phase a large number of spectral components.

Time-bandwidth relation



$\Delta\tau \,.\, \Delta\nu = K$

K = 0.44: Gaussian pulse

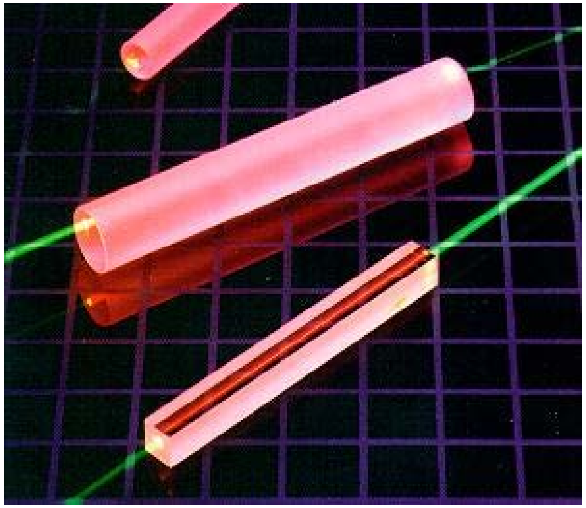
K=0.315 : sech^2 pulse

Need for large bandwidth amplifiers

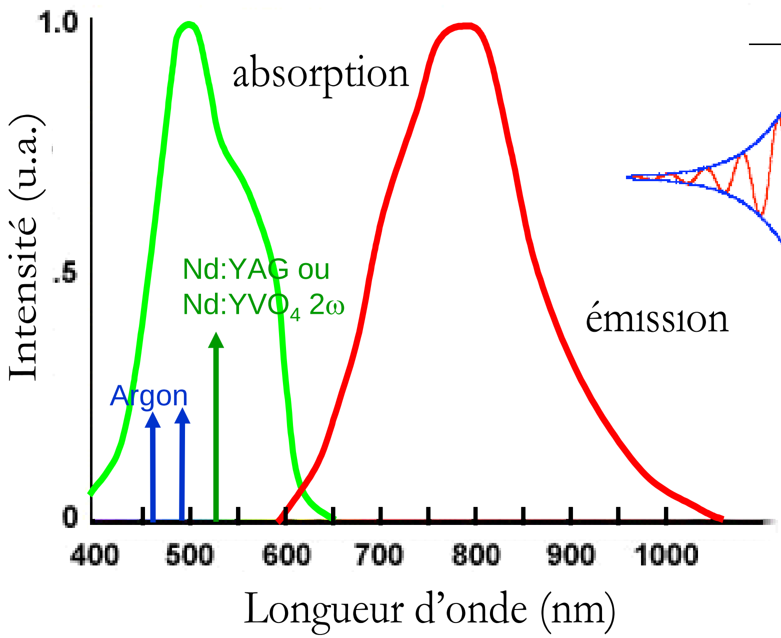
Best performances with Titanium-doped sapphire crystals $Ti^{3+}:Al2O_3$

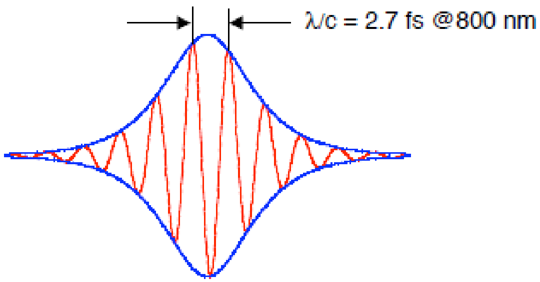
175 nm corresponds to 5,3 fs at 800 nm
≈ Duration ready available from Ti:Sa oscillators based

NB : 1 optical cycle



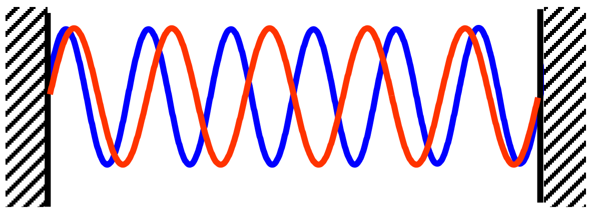
Ti3+ doping concentration : 1 % weight

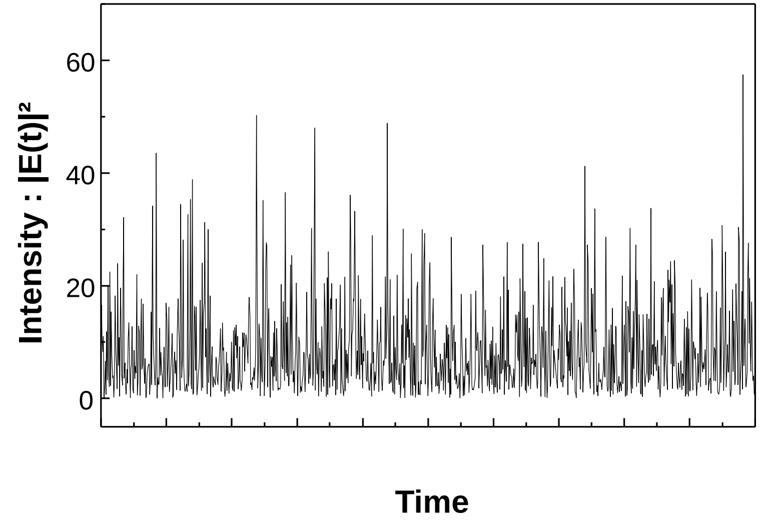




Ultrashort pulses generation – Mode-locking

Random phase modes :

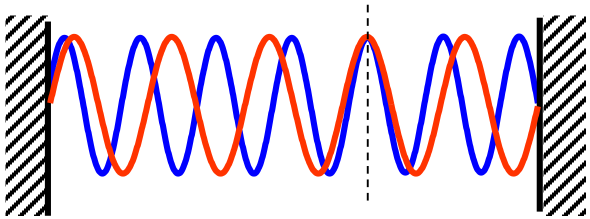


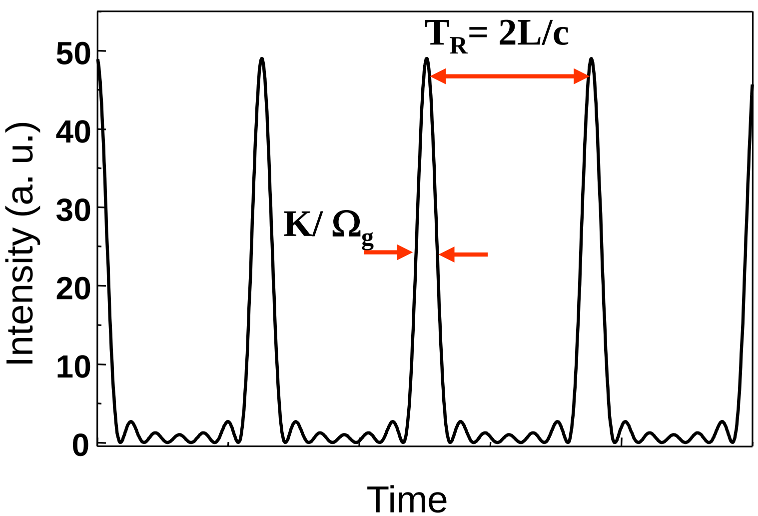


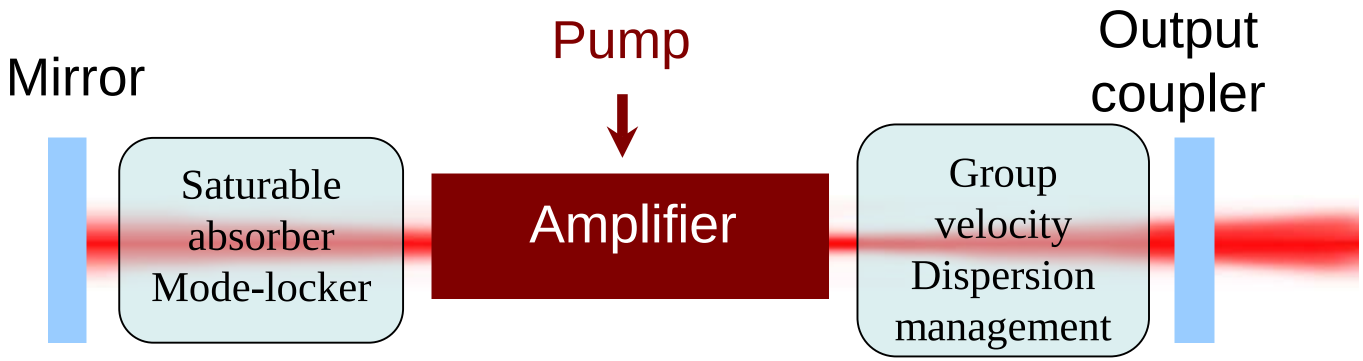
In phase modes

$\Phi_m = 0, \forall m$ $\Rightarrow$

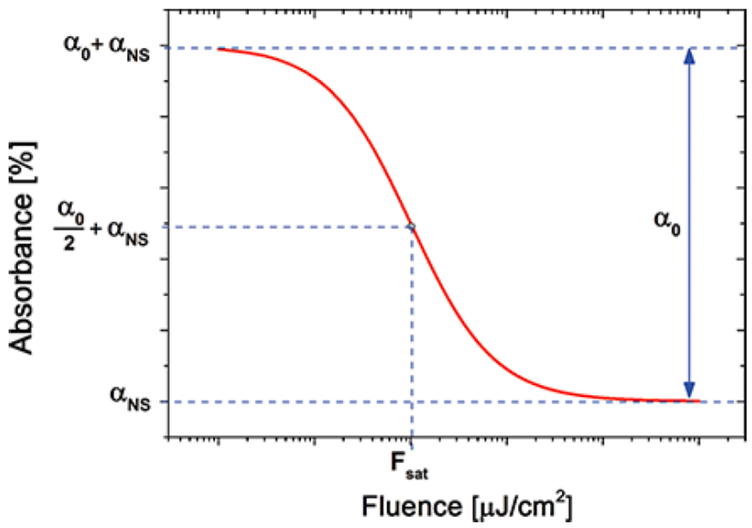
$$I(t) = I_0 \frac{\sin(N\omega_{isl} t/2)}{\sin(\omega_{isl} t/2)}$$







- Saturable absorber : promote pulsed operation against cw.
- High-intensity spikes burn through; low-intensity light is absorbed.

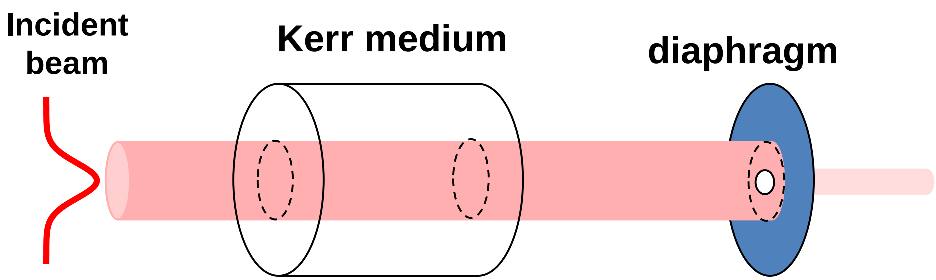


Several saturable absorbers:

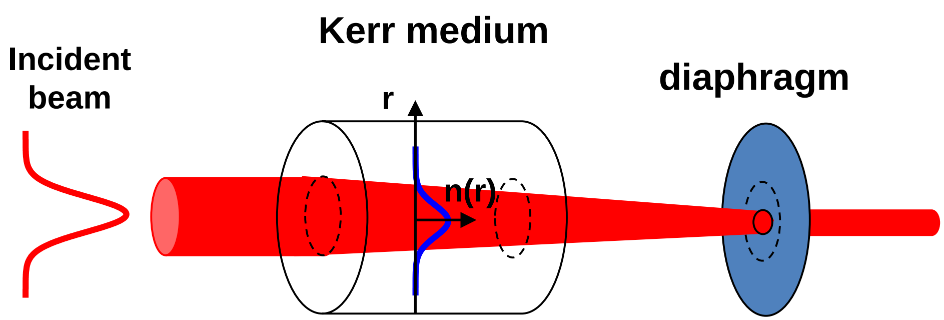
- Dye saturable absorbers
- Semiconductors (SESAMs)
- Graphene and carbon nanotubes
- Kerr-based saturable absorbers
- …

Ultrashort pulses generation – Mode-locking

Kerr-lens mode-locking (KLM)



Low intensity : low transmission

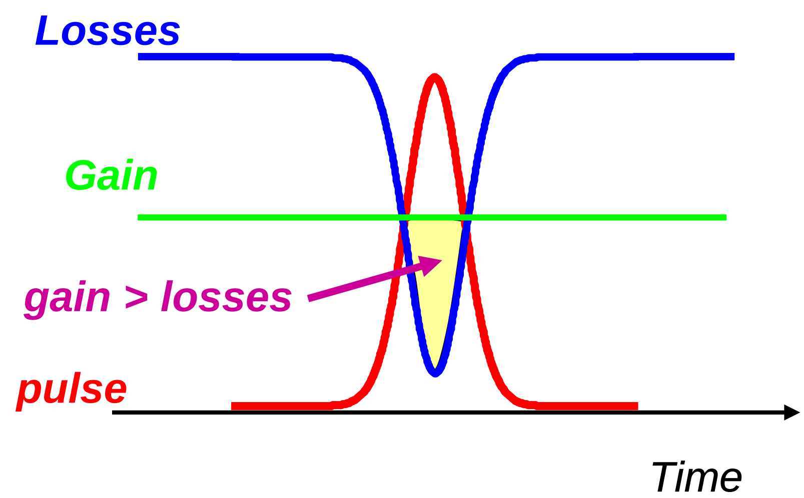


High intensity : high transmission

D. E. Spence, P. N. Kean, W. Sibbett, Opt. Lett. 16, 42, 1991

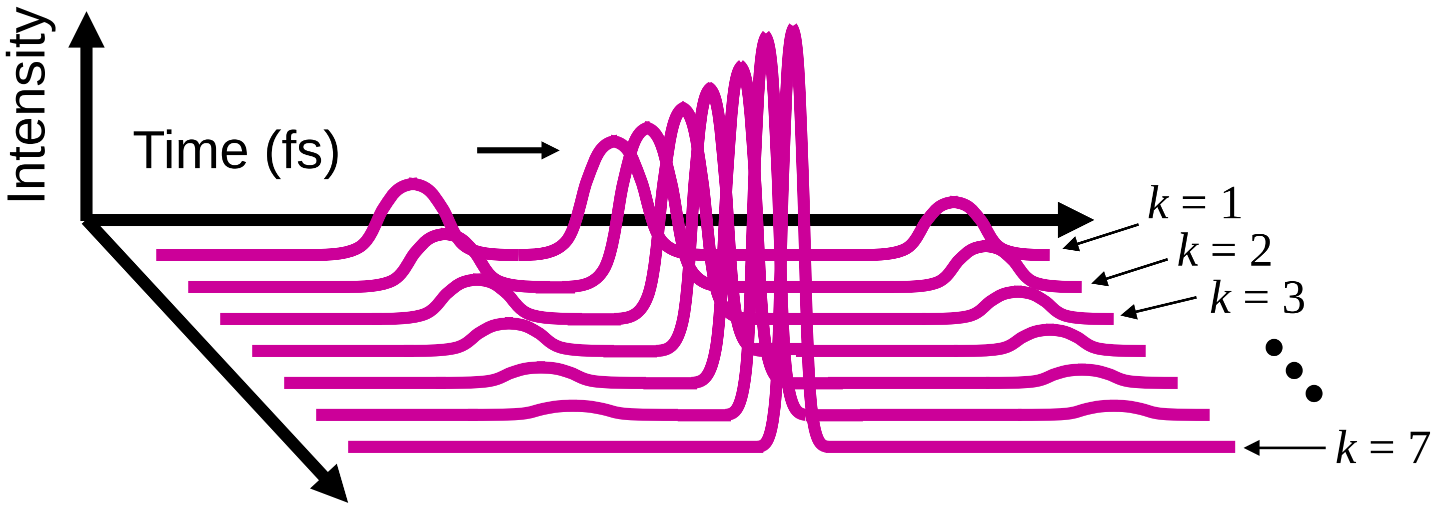
- **refractive index of the medium varies with intensity : $n(I) = n_0 + n_2 I$**

Fast saturable absorber



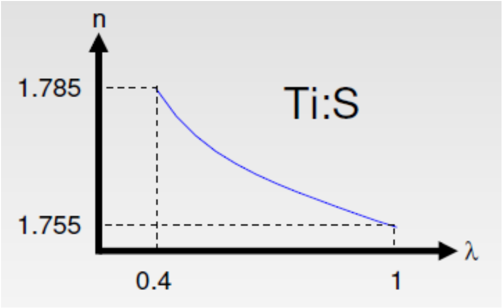
The pulse construct from noise (ns-ps):

- **The SA imposes high losses for low intensity structures**
- **The high-intensity noise structure is shortened after several round-trips**
- **Intermodals coherence constructs naturally!**



Propagation of broadband pulses ⇔ group velocity dispersion

- Refractive index varies with frequency



$$k(\omega) = n(\omega)\frac{\omega}{c}$$

$$k(\omega) = k_0 + \left.\frac{\partial k}{\partial \omega}\right|_{\omega_0}(\omega-\omega_0) + \frac{1}{2}\left.\frac{\partial^2 k}{\partial \omega^2}\right|_{\omega_0}(\omega-\omega_0)^2 + \frac{1}{6}\left.\frac{\partial^3 k}{\partial \omega^3}\right|_{\omega_0}(\omega-\omega_0)^3 + \ldots$$

$\equiv 1/\upsilon_g$ — **Group velocity**

$\equiv \beta_2\left[ps^2/m\right]$ — **2nd order GVD**

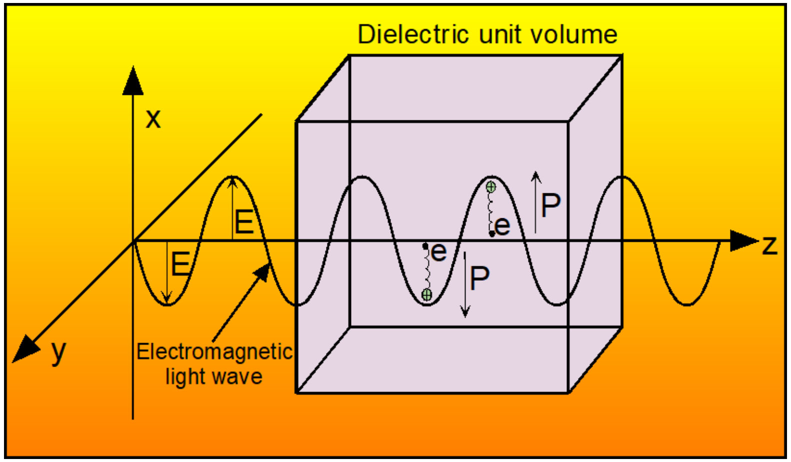
$\equiv \beta_3\left[ps^3/m\right]$ — **3rd ordre dispersion**

Accumulated phase:

$$\varphi(\omega) = k(\omega).\mathrm{d} = \varphi_0 + \varphi^{(1)}(\omega-\omega_0) + \varphi^{(2)}(\omega-\omega_0)^2 + \cdots$$

Propagation of intense pulses : nonlinear polarization component induced in the medium

$$P = \varepsilon_0(\chi^{(1)}E + \chi^{(2)}E^2 + \chi^{(3)}E^3 + \ldots)$$

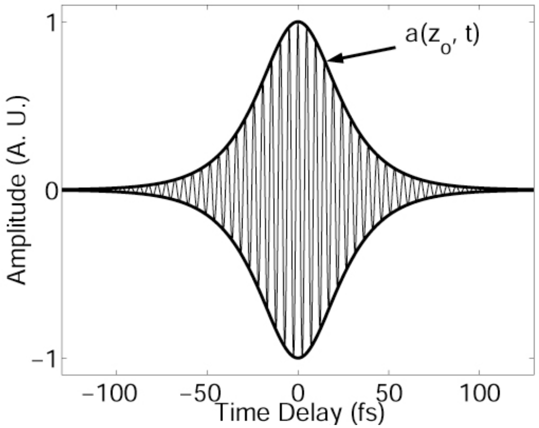


$\chi^{(1)}$	~~$\chi^{(2)}$~~	$\chi^{(3)}$
Linear susceptibility	~~**2^{nd} order**~~	**3rd order**
↓	↓	↓
Classical optical effects (reflection, absorption)	~~SHG, parametric mixing, electro-optics effect~~	Brillouin and Raman, Optical Kerr effect

$$\frac{\partial^2 \vec{E}(z,t)}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2 \vec{E}(z,t)}{\partial t^2} = \mu_o \frac{\partial^2}{\partial t^2}\left(\vec{P}_L(z,t) + \vec{P}_{NL}(z,t)\right)$$

$$\vec{P}_{NL}(z,t) = \varepsilon_o \chi^{(3)} \vdots E(z,t)E(z,t)E(z,t)$$

$$E(z,t) = c \cdot a(z,t) \cdot \exp(i\beta_0 z - i\omega_0 t)$$



- **Nonlinear Schödinger equation**

Describes the evolution of the pulse envelop in function of time and distance.

$$\frac{\partial a(z,t)}{\partial z} = -i\frac{\beta_2}{2}\frac{\partial^2 a(z,t)}{\partial t^2} + i\gamma\,|a(z,t)|^2\,a(z,t)$$

$$\gamma = \frac{n_2\omega_0}{cA_{eff}} > 0$$

Kerr coefficient with A_{eff} the area of the beam cross section

Time coordinate is shifted to eliminate the propagation delay

$$t = t_{\text{old}} - z/v_{\text{g}}$$

C. R. Menyuk, IEEE J. Quantum Electron. 25, 12, 2674-2682, December 1, 1989.
L. F. Mollenauer *et al.*, in *Opt. Fiber Telecommunications iVA*, (Academic, San Diego, Calif., 1997).

Linear pulse propagation – nonlinear Schrödinger equation

Low intensity : neglect nonlinear term (|a|² is low)

$$\frac{\partial a}{\partial z} = -i\frac{\beta_2}{2}\frac{\partial^2 a}{\partial t^2} + i\gamma|a|^2 a$$

Trivial solution in frequency domain :

$$\frac{\partial \tilde{a}}{\partial z} = i\frac{\beta_2}{2}\Omega^2\tilde{a}$$

$$\Omega = \omega - \omega_0$$

$$\tilde{a}(\Omega, z) = \exp(i\beta_2\Omega^2 z/2)\,\tilde{a}(\Omega, 0)$$

Multiplication by a quadratic phase term : power spectrum remains unchanged

UMR 6614

Temporal domain | **Frequency domain**

$$a(t,0) = A_0 \exp\left[-\frac{t^2}{2\tau_0^2}\right] \quad \xrightarrow{\text{FT}} \quad \tilde{a}(\Omega,0) = A_0\sqrt{2\pi\tau_0^2}\, e^{-\frac{\Omega^2}{2}\tau_0^2}$$

$$\times \exp(i\beta_2\Omega^2 z/2)$$

$$a(t,z) = A(z) e^{-\frac{1+iC(z)}{2\tau(z)^2}t^2} \quad \xleftarrow{\text{IFT}} \quad \tilde{a}(\Omega,z) = A_0\sqrt{2\pi\tau_0^2}\, e^{-\frac{\Omega^2}{2}(\tau_0^2 - i\beta_2 z)}$$

where :

$$L_D = \tau_0^2 / |\beta_2|$$ **Dispersion length**

$$\tau(z) = \tau_0\sqrt{1 + z^2 / L_D^2}$$ **Pulse width**

$$C(z) = \text{sign}(\beta_2) z / L_D = \beta_2 z / \tau_0^2$$ **Chirp parameter**

coRia UMR 6614

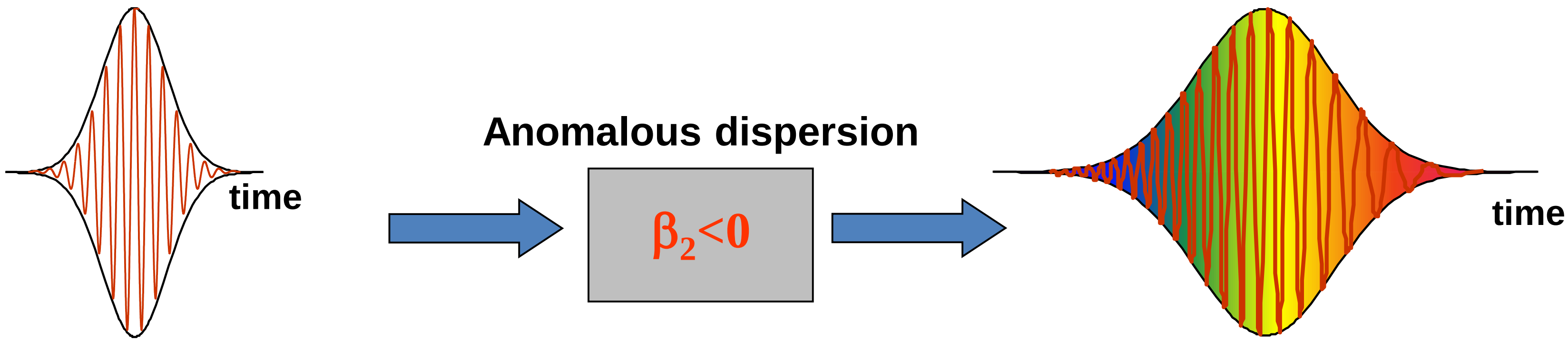
$$a(t,z) = A(z)\exp\left[-\frac{t^2}{2\tau(z)^2} + i\varphi_L(z,t)\right]$$

Parabolic phase : $\varphi_L = -\frac{C(z)}{2\tau(z)^2}t^2$

$\Rightarrow$ **Linear evolution of instantaneous frequency :** $\delta\omega_L(t,z) = -\frac{\partial\varphi_L}{\partial t} = \mathrm{sign}(\beta_2)\frac{z/L_D}{\tau(z)^2}t$

- **Temporal domain : pulse stretching**

The blue components propagate faster and arrive first

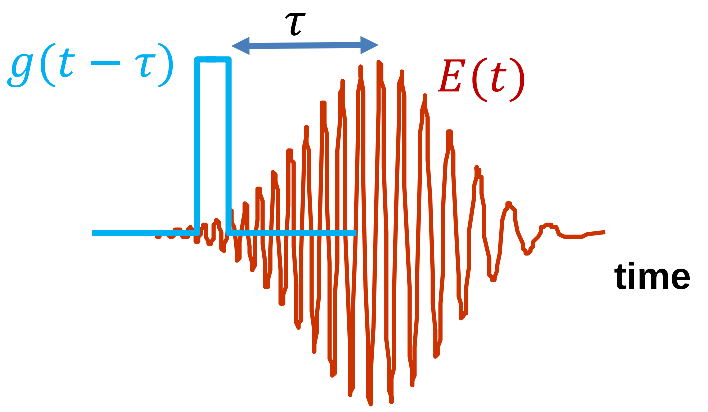


- **Spectral domain : the spectrum remains unchanged**

Frequency Resolved Optical Gating (FROG)

→ Measure the spectrogram given by :

$$S(\omega,\tau) = \left| \int_{-\infty}^{+\infty} E(t)\, g(t-\tau)\, e^{i\omega t}\, dt \right|^2$$



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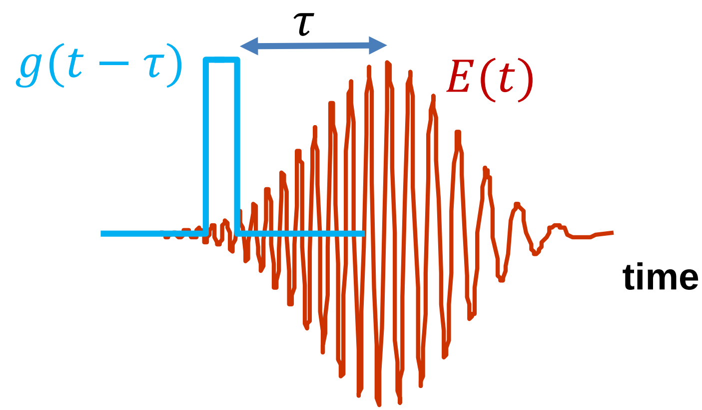
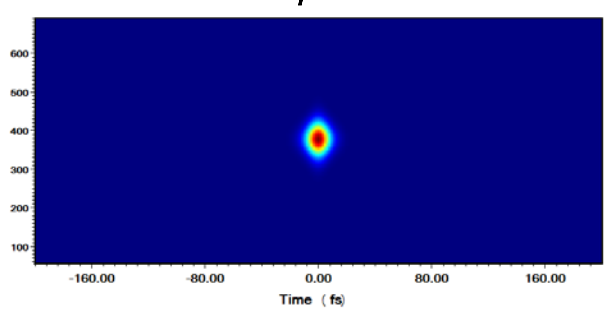
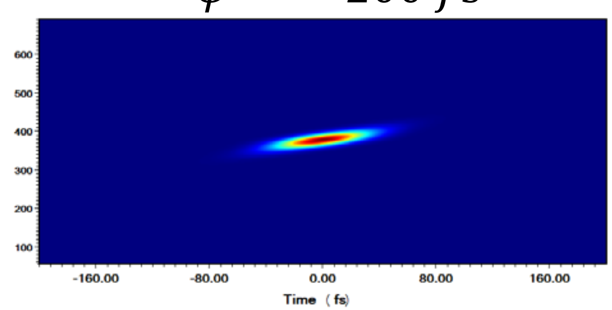


Spectrograms of a 10 fs pulse @ 800 nm

$\varphi^{(2)} = 0$



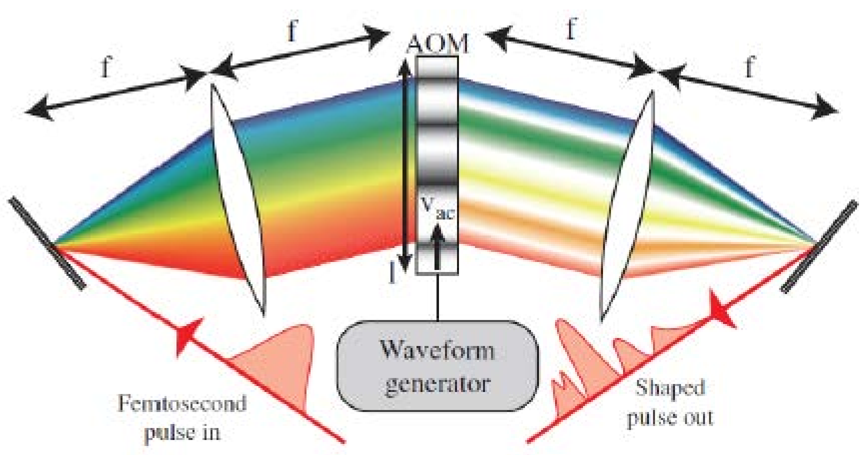
$\varphi^{(2)} = 200\ fs^2$



→ Iterative algorithms to retrieve the spectral phase distribution $\varphi(\omega)$

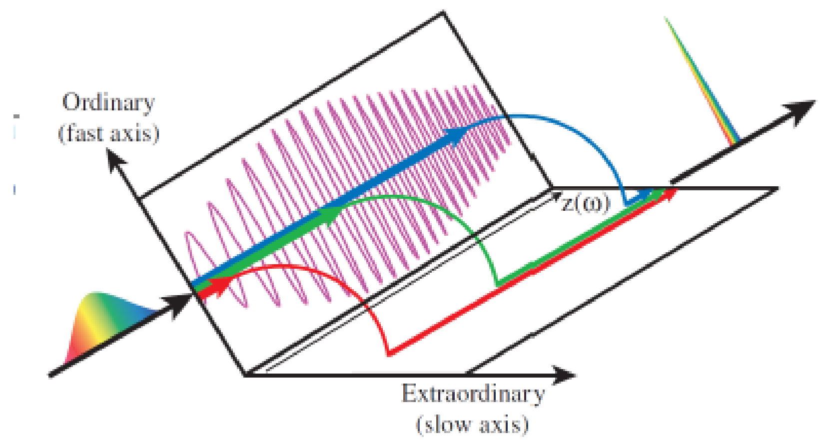
Modify the frequency phase to control the temporal shape

4f phase shaper



Discrete shaping of the phase by the AOM placed in the Fourier plane.

Dazzler



Shaping of spectral phase through chirp of acoustic wave.

Montmayrant & Blanchet, *J. Phys. B: At. Mol. Opt. Phys.* **43** 103001 (2010)

Short propagation length : we neglect the dispersion term

$$\frac{\partial a}{\partial z} = -i\frac{\beta_2}{2}\frac{\partial^2 a}{\partial t^2} + i\gamma|a|^2 a$$

$$\Downarrow$$

$$\frac{\partial a}{\partial z} \approx +i\gamma|a|^2 a(t,z)$$

Approached solution can be found assuming a constant power over dz :

$$\frac{\partial |a(t,z)|^2}{\partial z} = 0 \quad \Rightarrow \quad |a(t,z)|^2 = |a(t,0)|^2$$

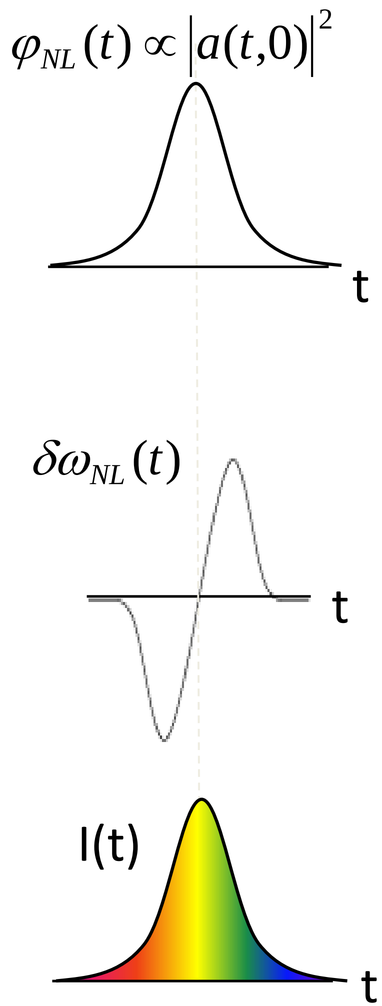
$$\Rightarrow \quad \boxed{a(t,z) = e^{+i\gamma|a(t,0)|^2 z} a(t,0)}$$

Nonlinear pulse propagation

$$a(t,z) = e^{+i\gamma|a(t,0)|^2 z}a(t,0) = e^{+i\varphi_{NL}(t,z)}a(t,0)$$

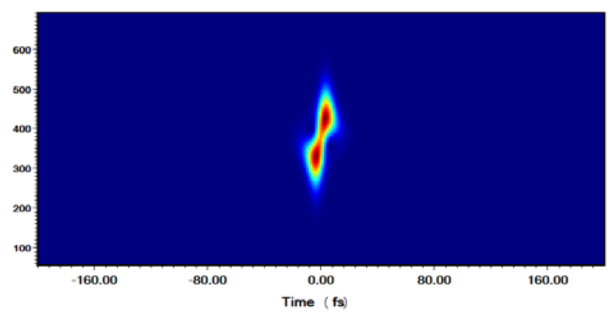
The pulse acquires chirp :

$$\delta\omega_{NL} = -\frac{\partial\varphi_{NL}}{\partial t} = -\gamma\frac{\partial|a(t,0)|^2}{\partial t}z = -\frac{2\pi n_2 z}{\lambda A_{eff}}\frac{\partial|a(t,0)|^2}{\partial t}$$

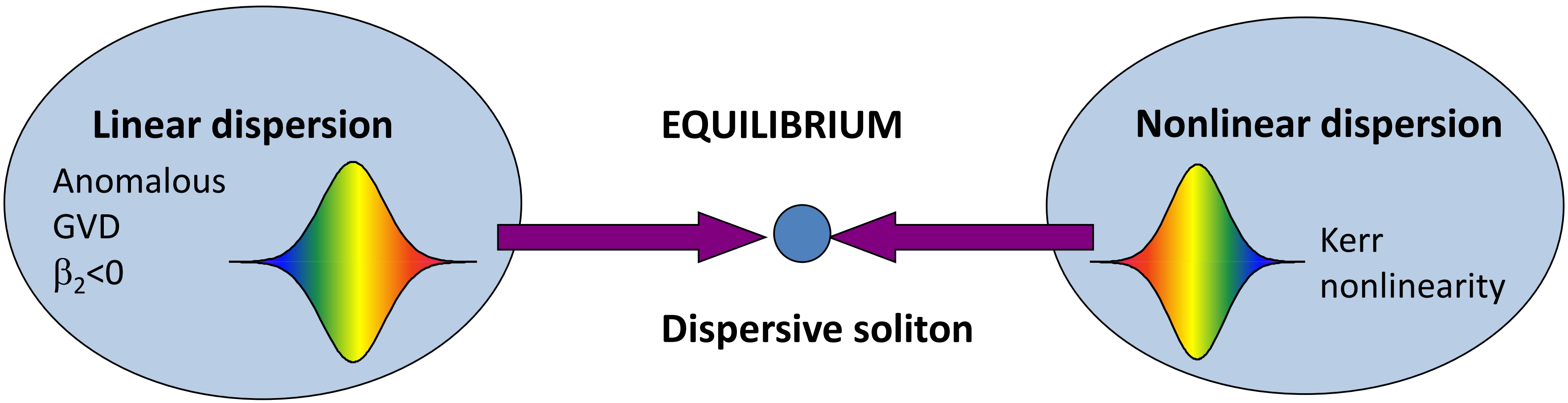


- ☞ ***New frequencies are created***
- ☞ ***For n_2>0 : frequencies distribution similar to normal dispersion.***
- ☞ ***Nonlinear chirp could be compensated by anomalous dispersion.***

Propagation along 2 m fiber



$$\frac{\partial a(z,t)}{\partial z} = -i\frac{\beta_2}{2}\frac{\partial^2 a(z,t)}{\partial t^2} + i\gamma\,|a(z,t)|^2\,a(z,t)$$



Dispersion & nonlinearity compensate exactly for an hyperbolic secant pulse profile :

$$a(t) = \mathrm{sech}(t/\tau_p)\exp(iz/z_{sol})$$

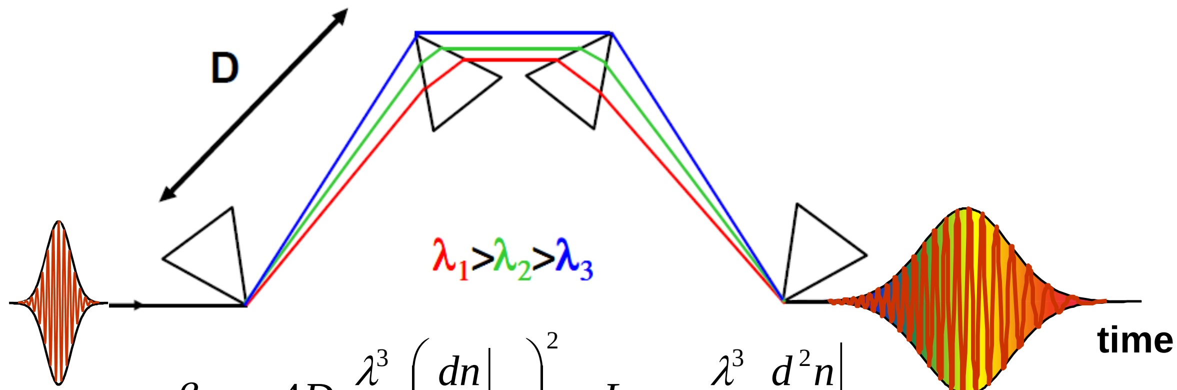
Ideal medium : homogeneous, isotropic and transparent!

Zakharov and Shabat, Sov. Phys. JETP 34, 62 (1972), Hasegawa and Tappert (1973), APL 23, 142 (1973)

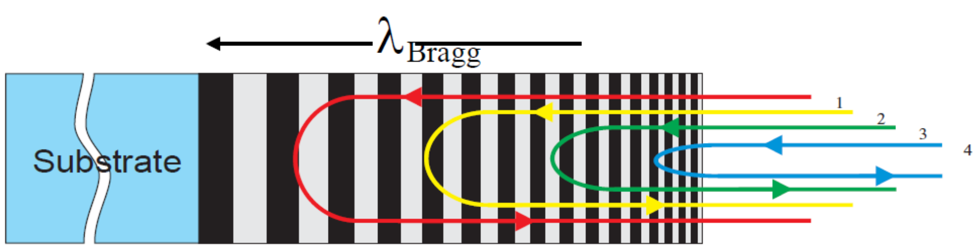
Ultrashort pulses propagation – Dispersion management

Glass materials : positive (normal) GVD in the visible and near infrared.

Dispersion management systems : prism pairs, grating pairs, chirped mirrors, GTI mirrors, chirped Bragg gratings.

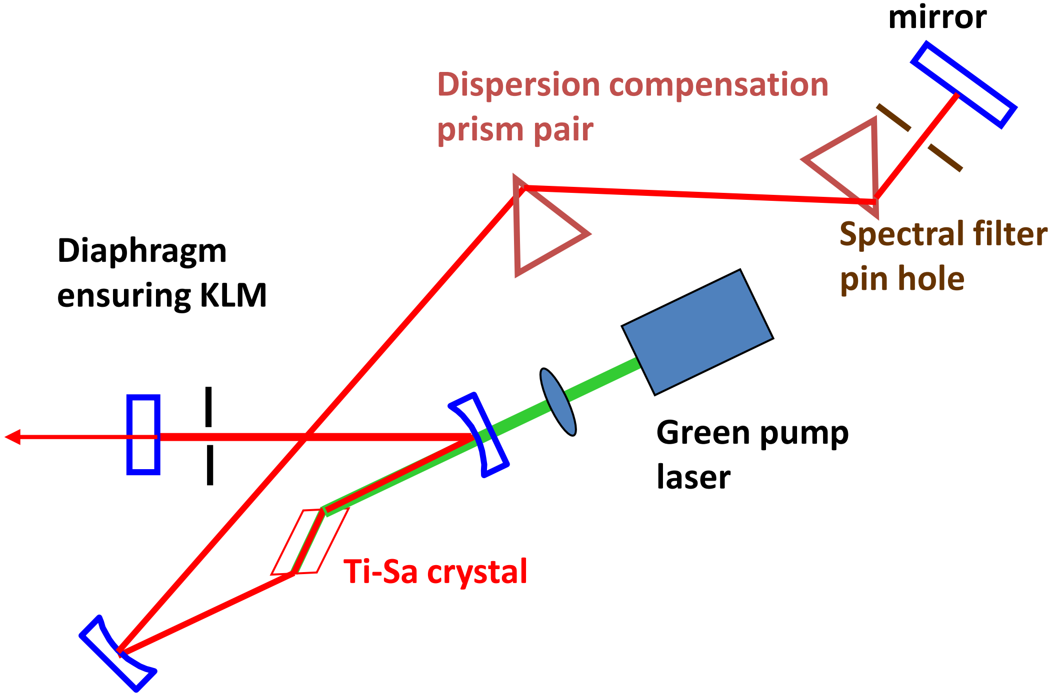


$$\beta_2 \approx -4D\frac{\lambda^3}{\pi c^2}\left(\left.\frac{dn}{d\lambda}\right|_{\lambda_0}\right)^2 + L_{prism}\frac{\lambda^3}{\pi c^2}\left.\frac{d^2n}{d\lambda^2}\right|_{\lambda_0}$$



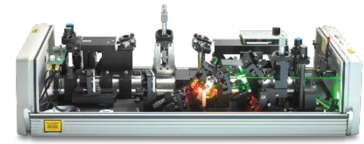
R. Szipöcs et al., Opt. Lett. 19, 201 (1994)

Mode-locked oscillators



Typical performances:
- Pulse duration < 100 fs
- Tunable from 700 nm to 1080 nm
- Energy = 10 nJ
- Repetition rate : MHz

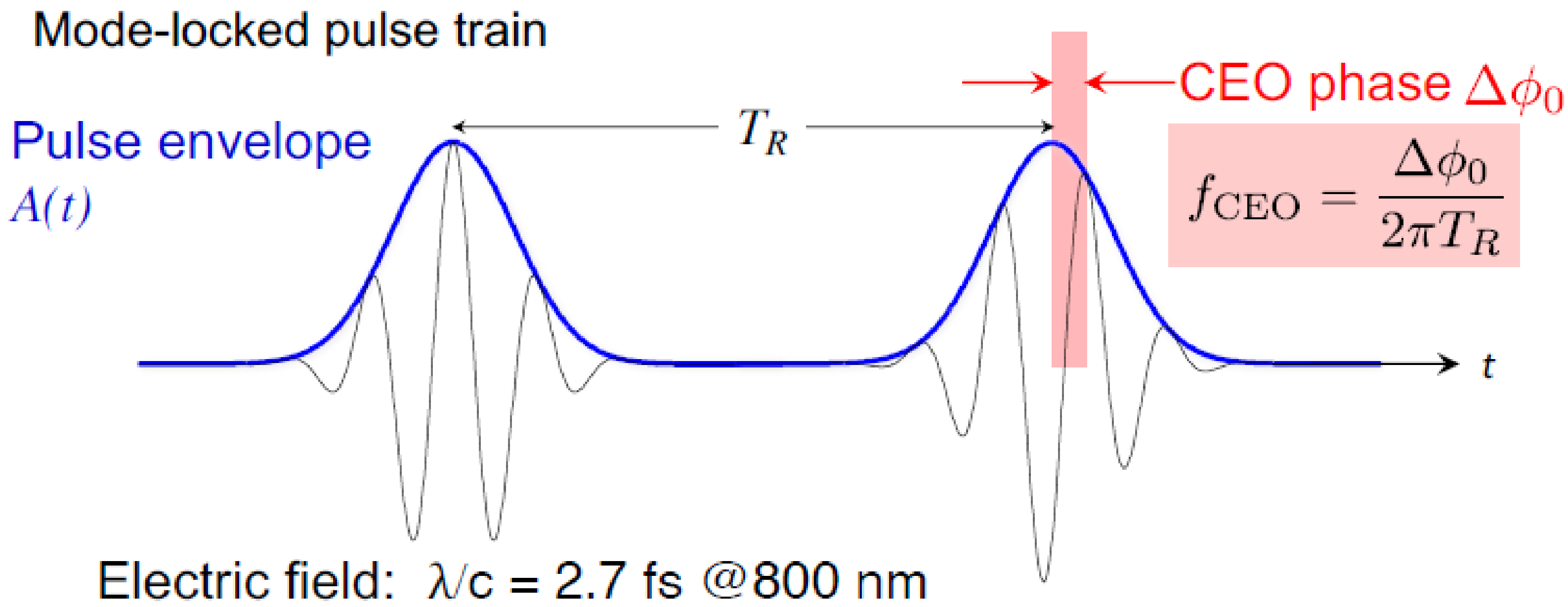
https://www.spectra-physics.com



https://fr.coherent.com

https://www.laserquantum.com

https://www.thorlabs.com



Need to stabilize repetition rate and carrier envelop offset phase

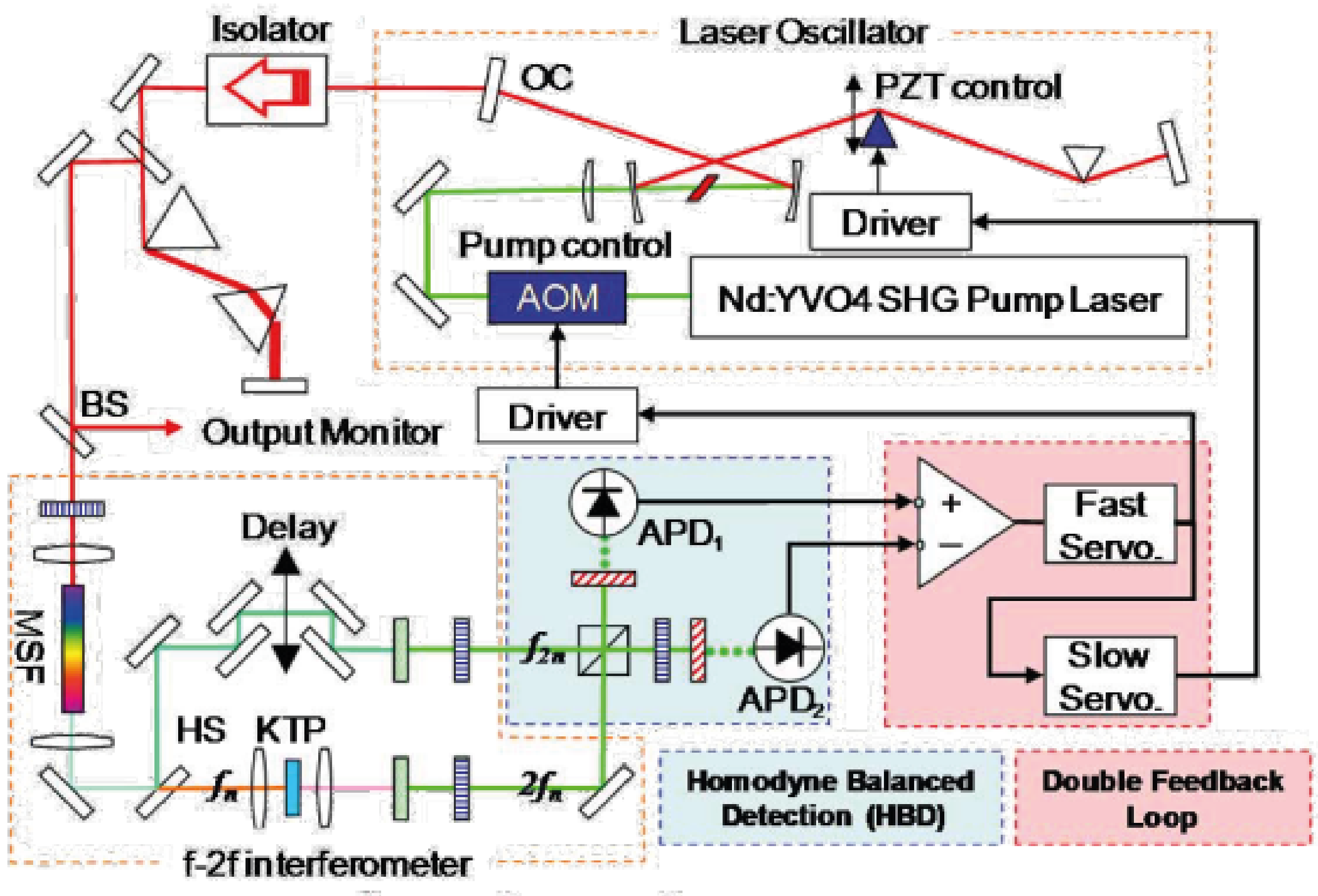
$$f_R = \frac{1}{T_R} = \frac{v_g}{2l_{las}}$$

Cavity length control

$$f_{CEO} = \left(1 - \frac{v_g}{v_\emptyset}\right) \cdot f_{las}$$

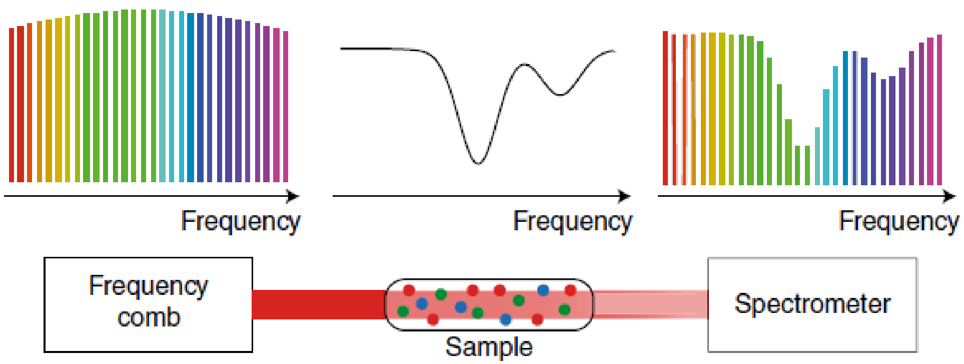
Phase and group velocities control

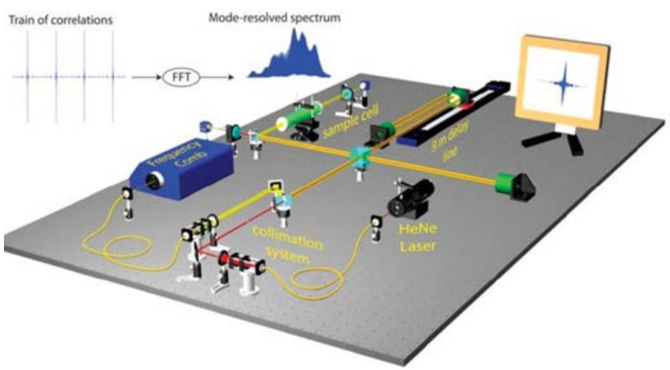
f-2f interferometer for CEO phase measurements



D. J. Jones et al. Science **288**, 635 (2000), A. Apolonski et al., Phys. Rev. Lett. **85**, 740 (2000)

- **Spectrum of a femtosecond laser pulse consists of millions of sharp lines**
- **These lines are equidistant across the entire spectrum**
- **A femtosecond laser is a “ruler” for frequencies !**



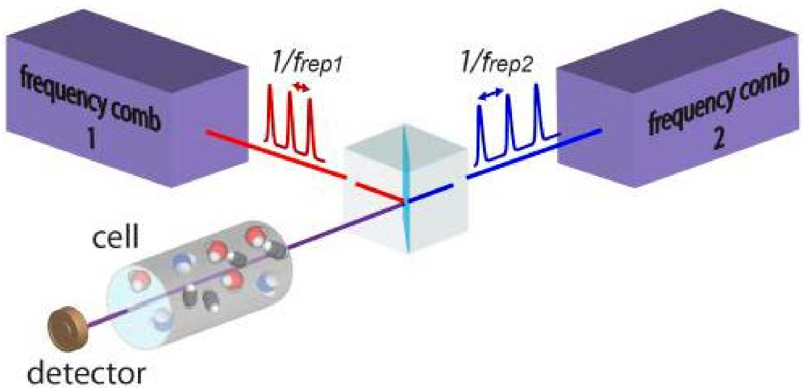


Prix Nobel 2005 – J. L. Hall et T. W. Hänsch

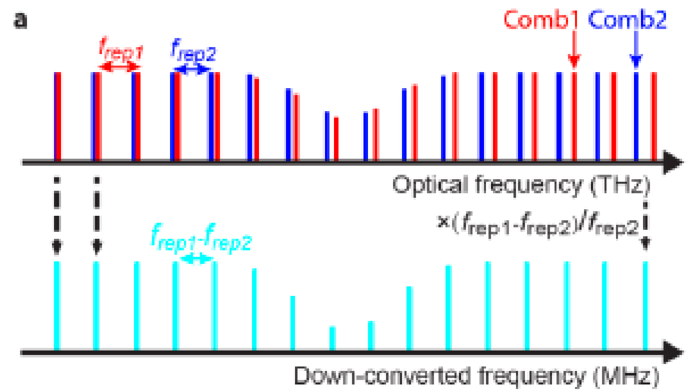
Need for FTIR or VIPA to resolve the comb components!

T. Udem, R. Holzwarth, T. W. Hänsch, *Nature* **416**, 233 (2002), N*ature Photonics* **volume 13**, pages146–157 (2019) , S. A. *Diddams et al.*, Nature, vol. 445 (2007),

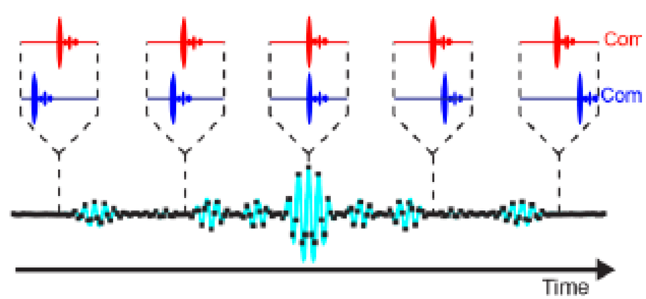
Dual comb spectroscopy



Frequency domain



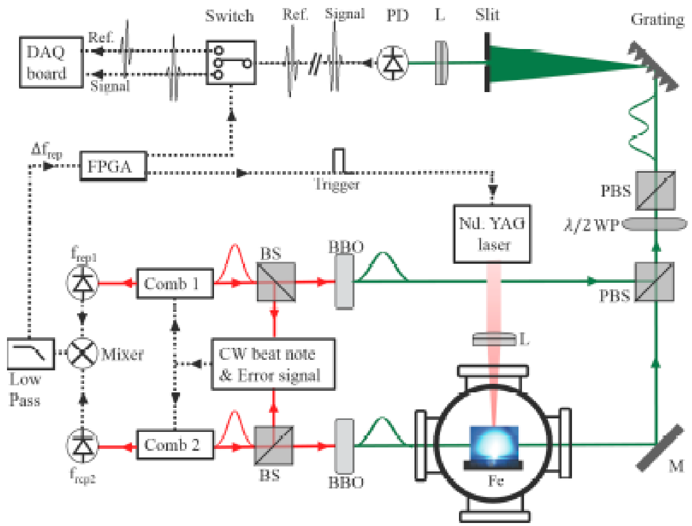
Time domain



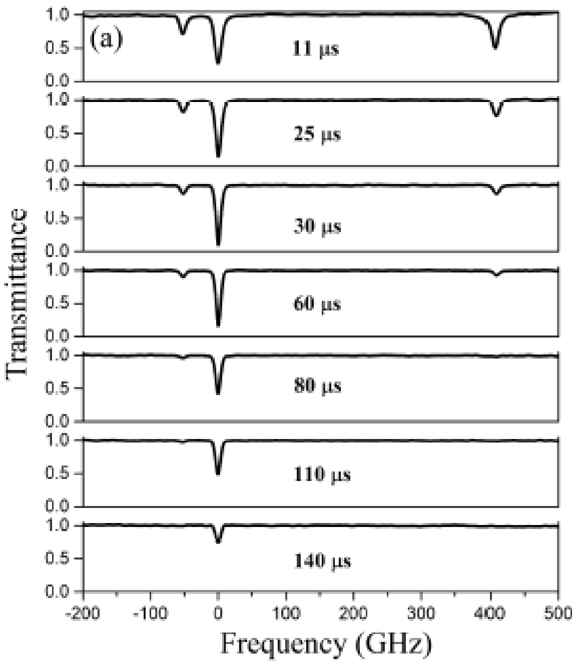
- Combine two optical frequency combs
- Intensity beat on photodetector
- Down-conversion to radio frequencies (RF)

T. H. Hänsch, N. Picqué, Jour. of Phys.: Conf. Series467 (2013), G. Millot en a;., Nat. Photon. **10**, 27–30 (2016)

Dual comb spectroscopy of laser induced plasma



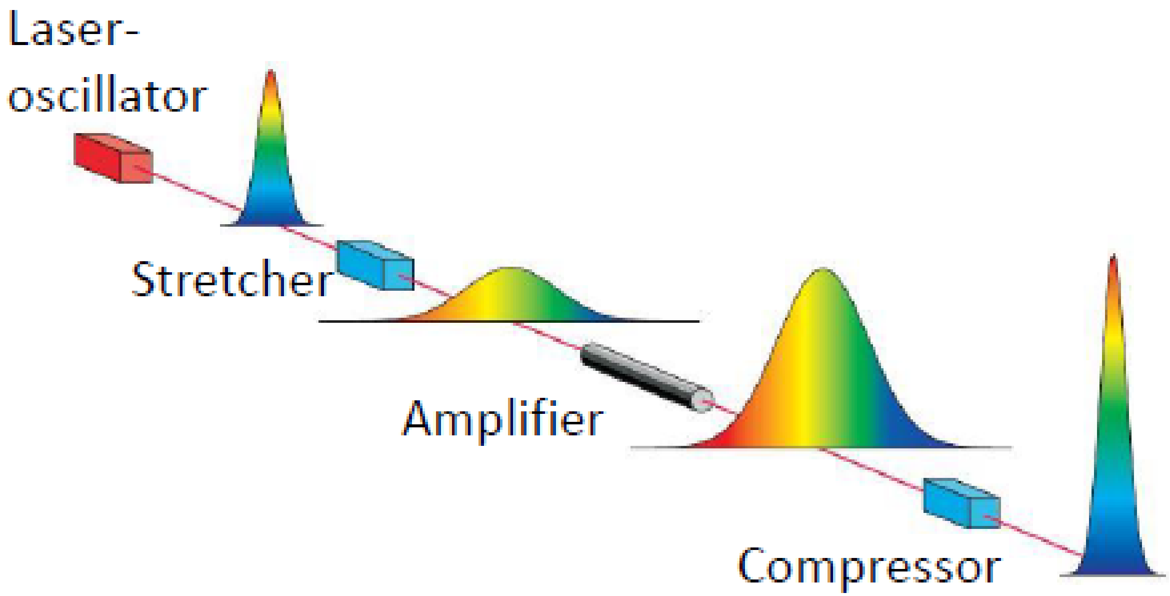
Stainless steel spectra at 533nm
Resolution : 8GHz (0.0076 nm)

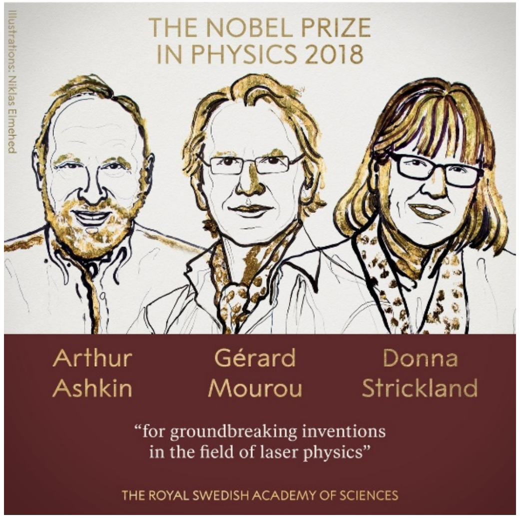


- The high spectral resolution and broad spectral coverage
- Time-resolved measurement capability

Y Zhang, C Lecaplain, RRD Weeks, J Yeak, SS Harilal et al., Optics letters, 2019

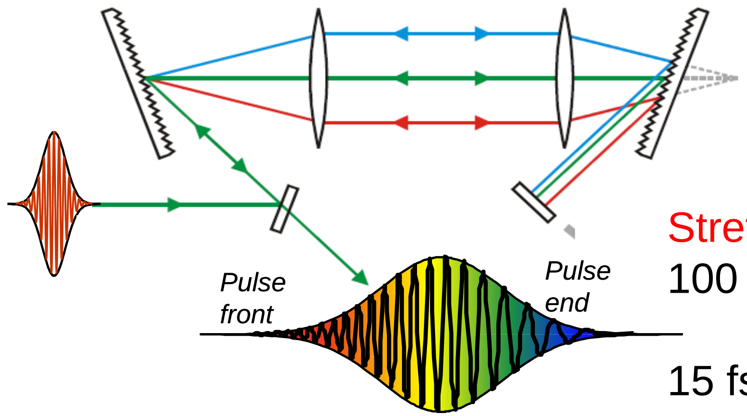
Energy scaling concept
Minimize impact of nonlinear effects





D. Strickland and G. Mourou, Opt. Commun. **56**, 219–221 (1985).

Need for large stretching ratios : grating-based stretcher

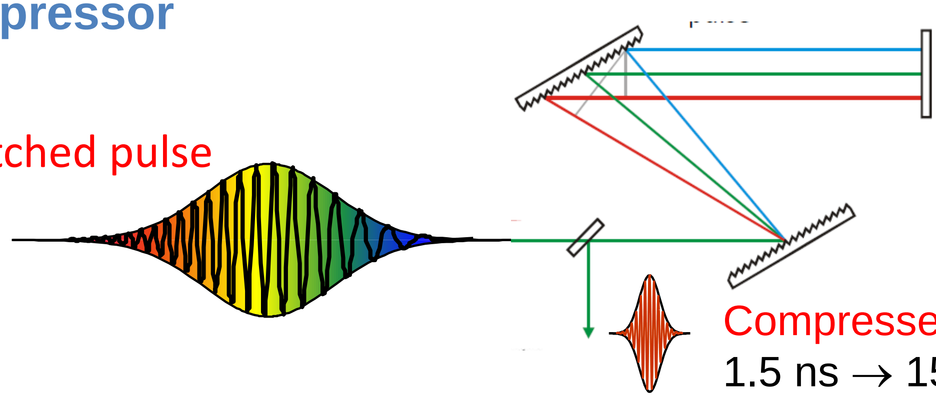


Stretched pulse
100 000 times!

15 fs → 1.5 ns

Grating-based compressor

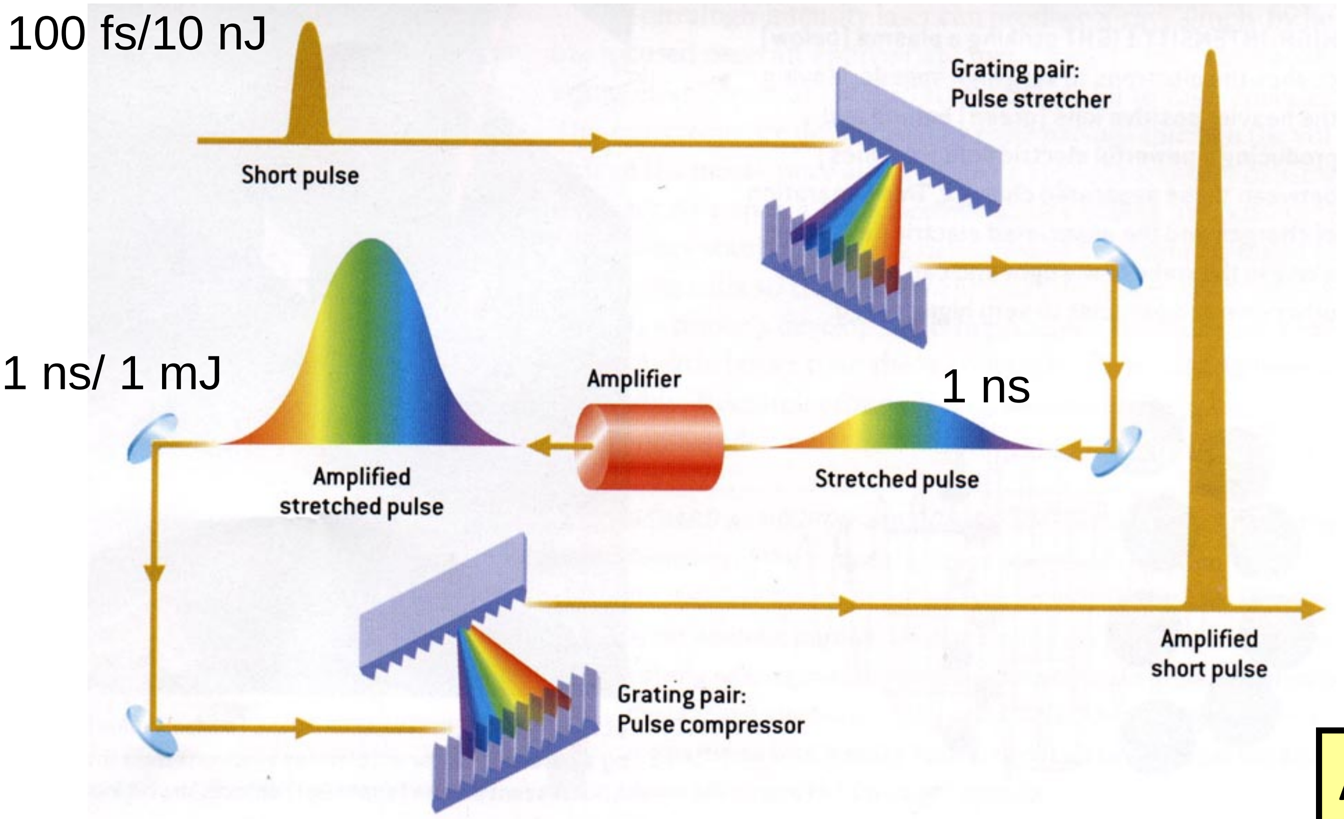
Stretched pulse



Compressed pulse
1.5 ns → 15 fs

E. B. Treacy, IEEE JQE 5, 454 (1969); Fork et al., Opt. Lett. 9, 150 (1984)

Amplification à dérive de fréquence (CPA)



$\Delta T = 100$ fs
E = 1mJ

Peak power :

$$P_{peak} = \frac{E}{\Delta T}$$

Focalisation to small spot « A »

$$I_{peak} = \frac{P_{peak}}{A}$$

A ~ 10 µm^2
$P_{crête}$ = 10 GW
$I_{crête}$ = 10^{17} W/cm^2

D. Strickland and G. Mourou, Opt. Commun. 56, 219–221 (1985), P. Maine et al., IEEE J. Quantum Electron. 24, 398 (1988).

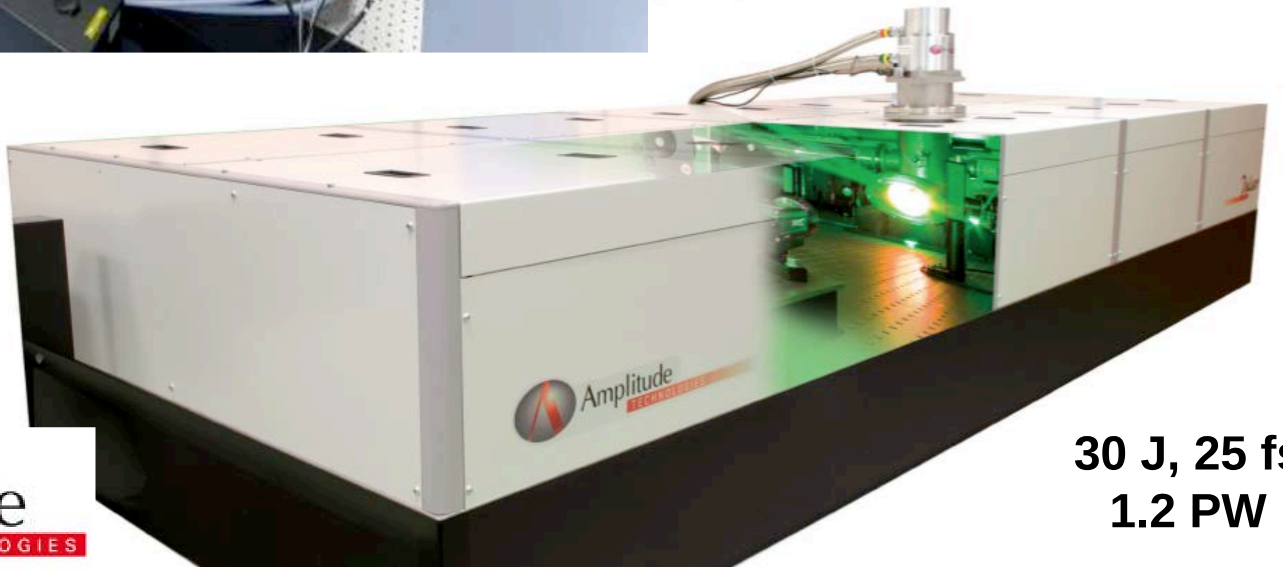
Standard commercial products

Ti-Sa lasers : 35 – 120 fs, multi-mJ @ 1 Khz

THALES

BELLA

40 J, 30 fs
1.3 PW



Amplitude TECHNOLOGIES

30 J, 25 fs
1.2 PW

Diode-pumped Ytterbium-doped lasers

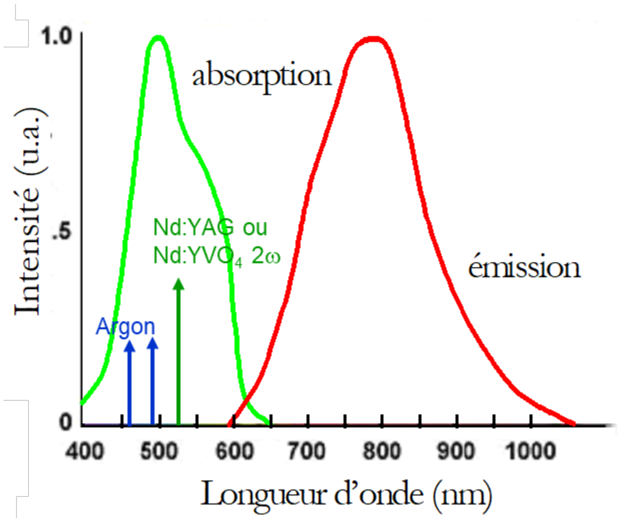
UMR 6614

Limitation of titanium-sapphire laser systems

☹ **Low efficiency : pumping in the green**

☹ **Thermal management**

☹ **Complexity and cost**

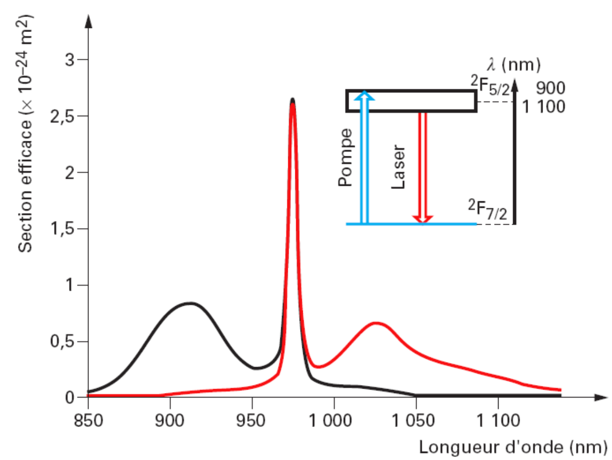


Ytterbium-doped host materials

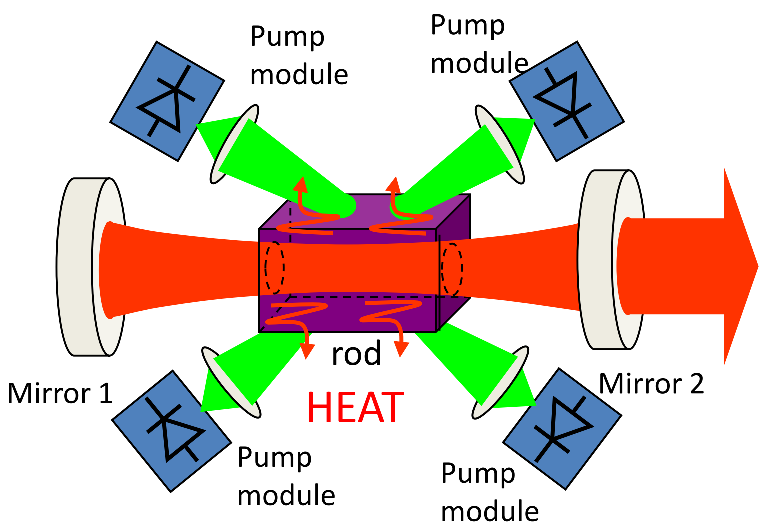
☺ **Low quantum defect**

- **Good thermal conductivity**
- **Large gain bandwidth**

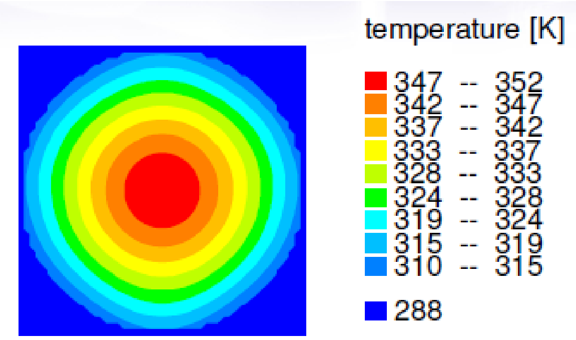
☺ **Diode pumping at 980 nm**



Conventional laser

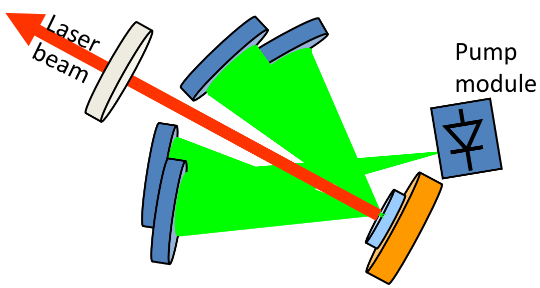


⇒ power dependent thermal lensing and thermal stress-induced birefringence

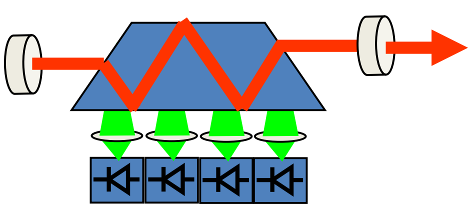


Solutions to reduce thermo-optical issues

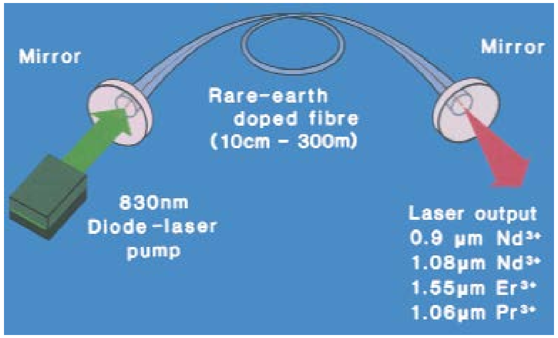
Thin-disk lasers



Slab lasers



Fibre lasers



Yb-lasers : 300- 500 fs, >100-µJ @ >100 kHz