Control of dispersion effects for resonant ultrashort pulses

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Context:

- Dispersion distorts the pulse. The sample is excited by a different field.
- A lot of physical and chemical processes depend on pulse temporal shape and phase

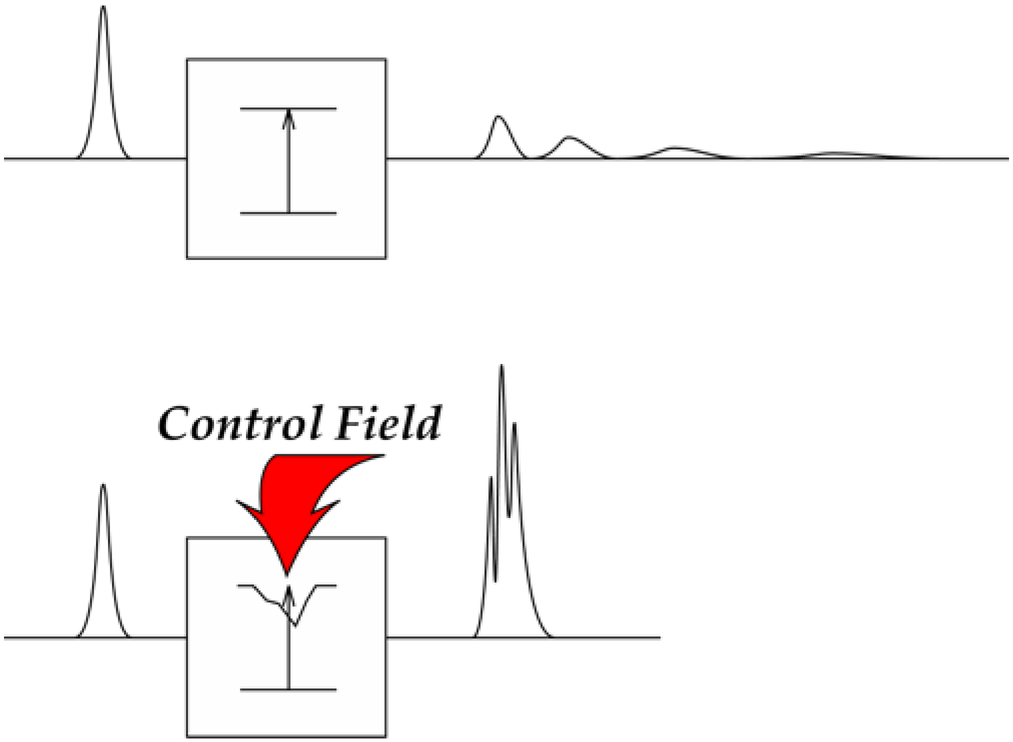
Shaping devices are limited :

- Wavelength
- Passive :No Amplification, Cannot create new frequencies

Resonant atomic dispersion and light-shifts may be an alternative

At atomic resonance:

- Gain
- Modification of pulse shape



1. Propagation of ultrashort pulses
2. Direct compensation with a pulse shaper
3. Case of an ultrashort pulse train.
4. Propagation in an atomic system driven by a strong pulse
5. Towards « active » pulse shaping

1. Propagation of ultrashort pulses

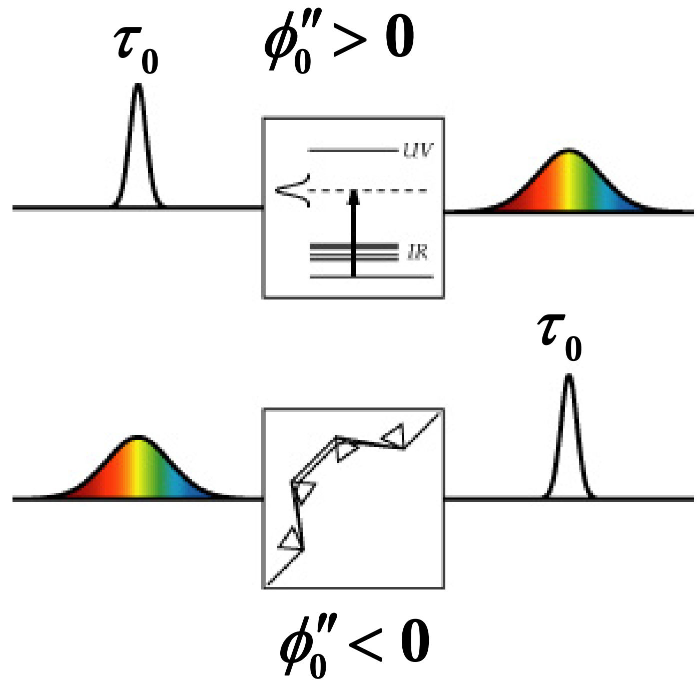
a) Non resonant medium

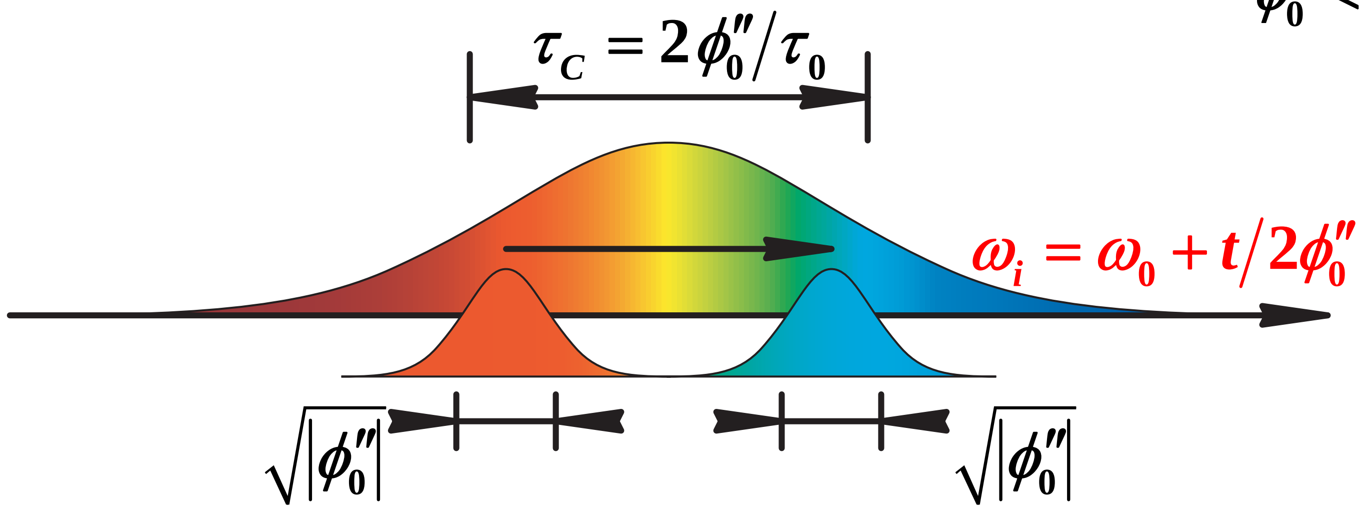
- Transparent

- Dispersion :

$$\phi(\omega) = n(\omega)\omega L/c$$

$$\phi(\omega) = \phi_0 + \phi_0'(\omega-\omega_L) + \phi_0''(\omega-\omega_L)^2/2$$





ω_0

1. Propagation of ultrashort pulses

b) Resonant (two level system)

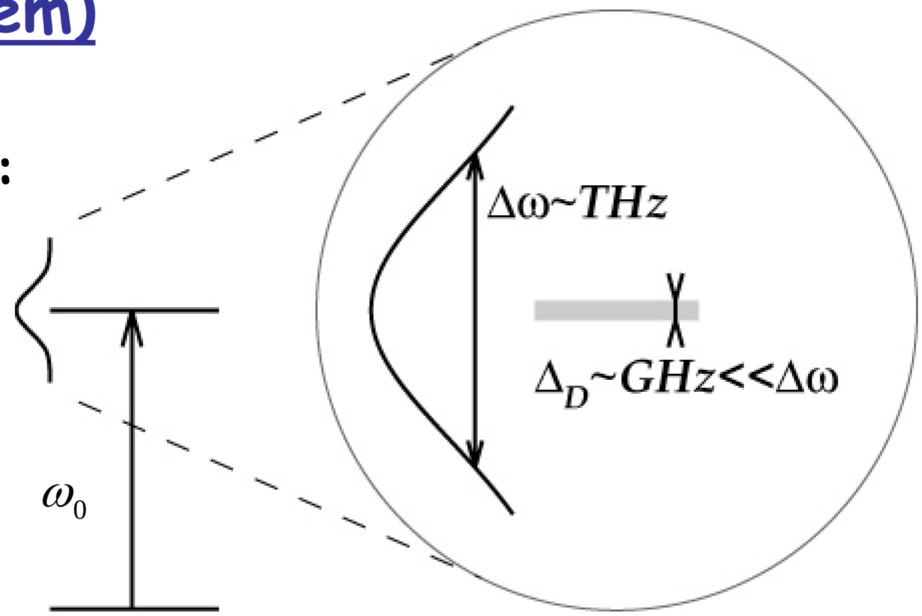
- **Total absorption negligible:**

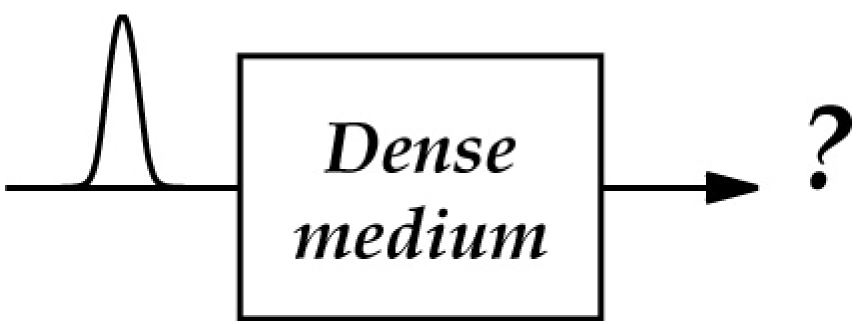
$$\Gamma << \Delta_D << \Delta\omega$$

- **Dispersion :**

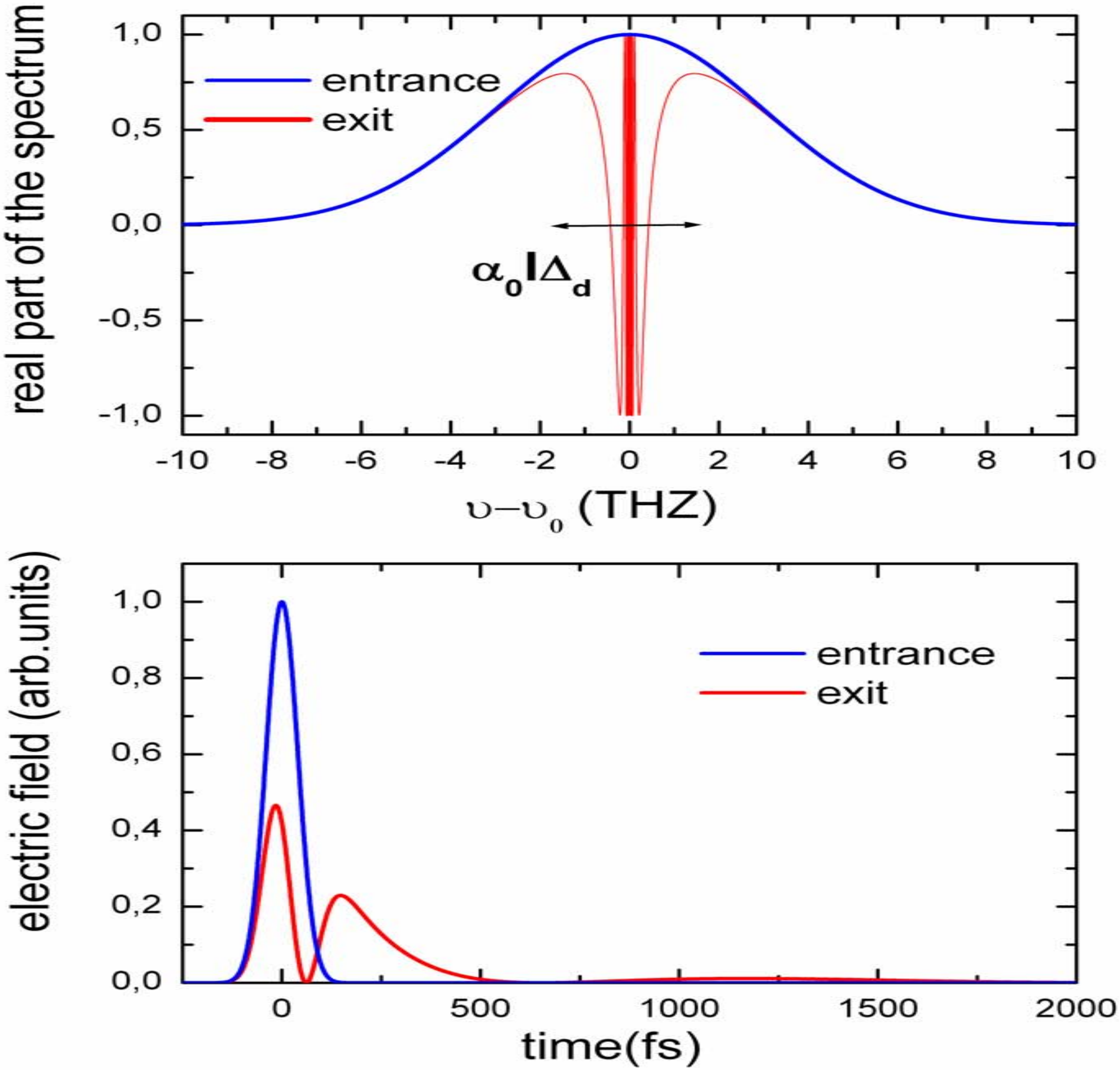
$$\phi(\omega) \approx \frac{\alpha_0 L \Delta_D}{\omega - \omega_0}$$

Optical depth





Rubidium, $4S_{1/2}$ -- $4P_{1/2}$ τ_0=75fs; $\alpha_0 l\Delta_d$=3THz;

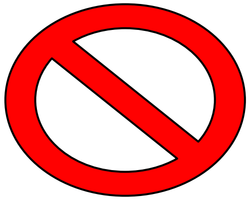


COMPENSATION ?

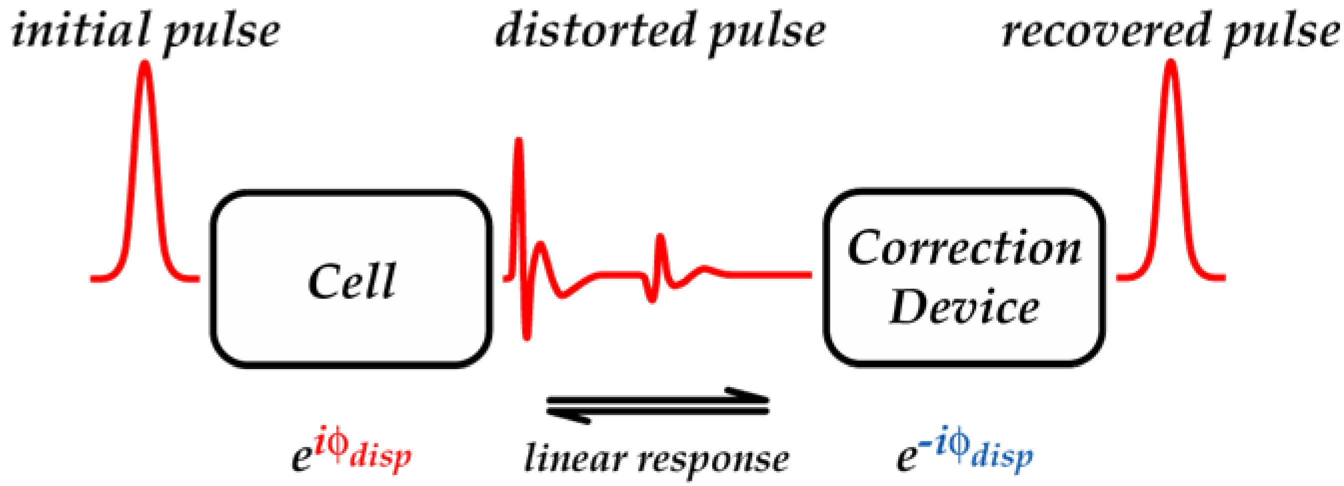
$$\phi(\omega) \simeq \frac{\alpha_0 L \Delta_D}{\omega - \omega_0}$$

Cannot be developed around
central laser frequency

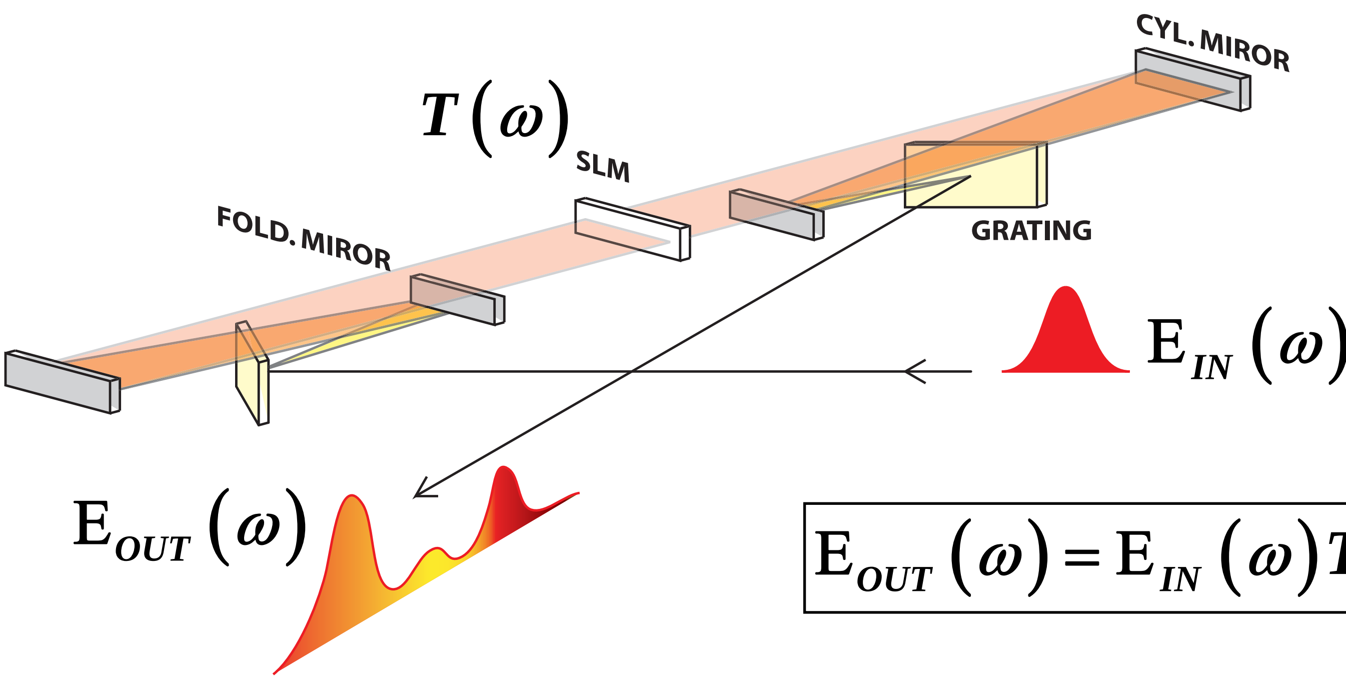
Second order no longer representative
All order are involved


Compensation with standard devices

2. Compensation with a pulse shaper

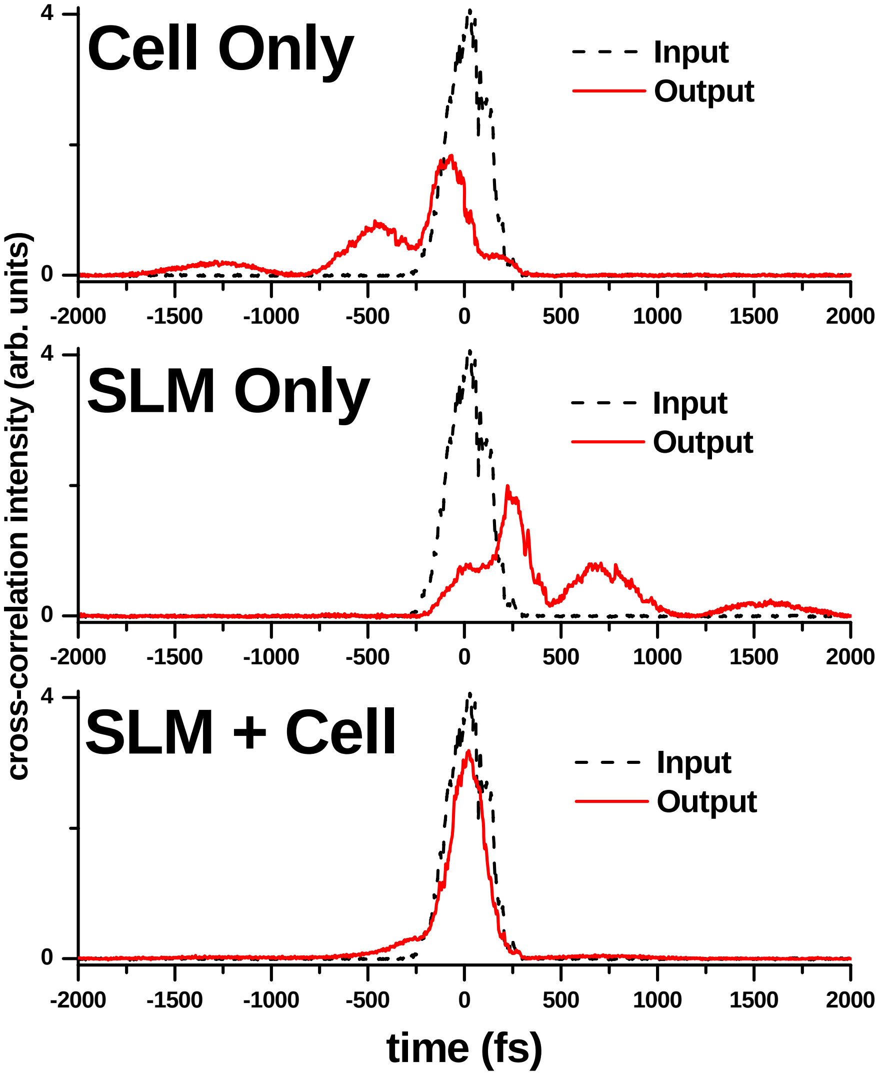


<u>640 pixels phase-amplitude SLM : 0,06 nm</u>



$$E_{OUT}(\omega) = E_{IN}(\omega)\,T(\omega)$$

Compensation with a pulse shaper



$$\tau_{FWHM} : 120\ fs, \lambda = 794,76\ nm$$

$$Rb : 5\,^2S_{1/2} \rightarrow 5\,^2P_{1/2}, \alpha_0 L \simeq 21500$$

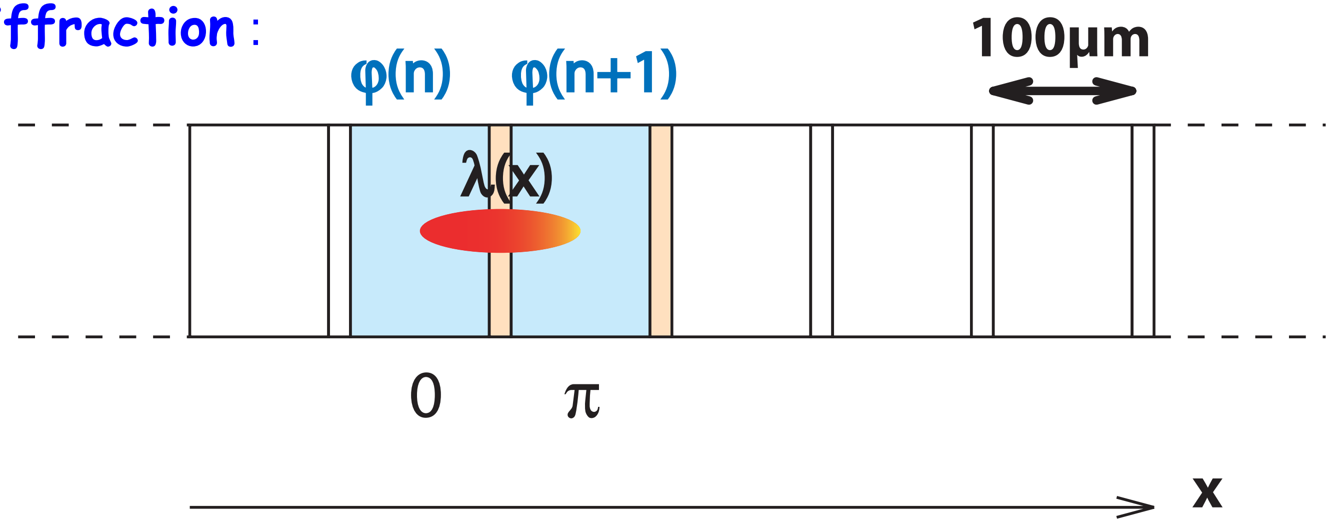
Efficient Compensation

Up to 85% of the incident energy recovered below the initial pulse envelop

Origin of Limitations:

1. Pixelisation: under-sampling (0.06 nm)

2. Diffraction :



Finite spot size for each spectral component

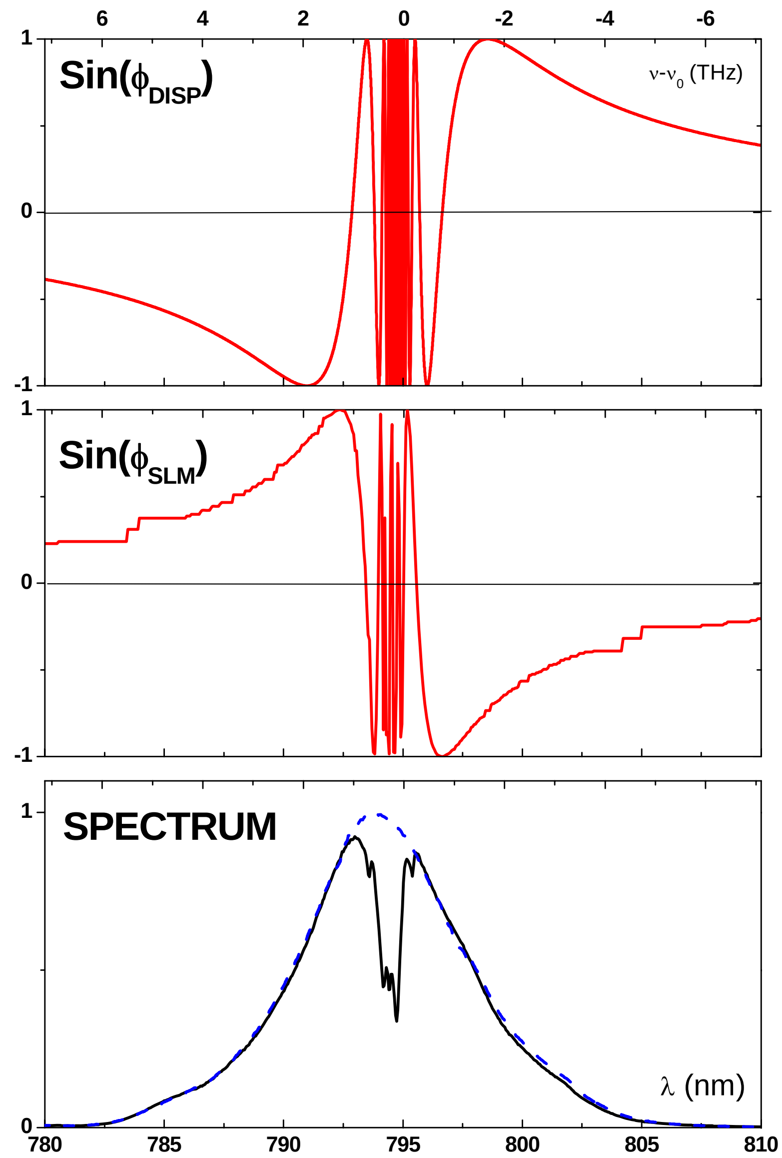
When Laser spot covers 2 **Pixels**

$$\varphi(n+1) \neq \varphi(n)$$: Interferences

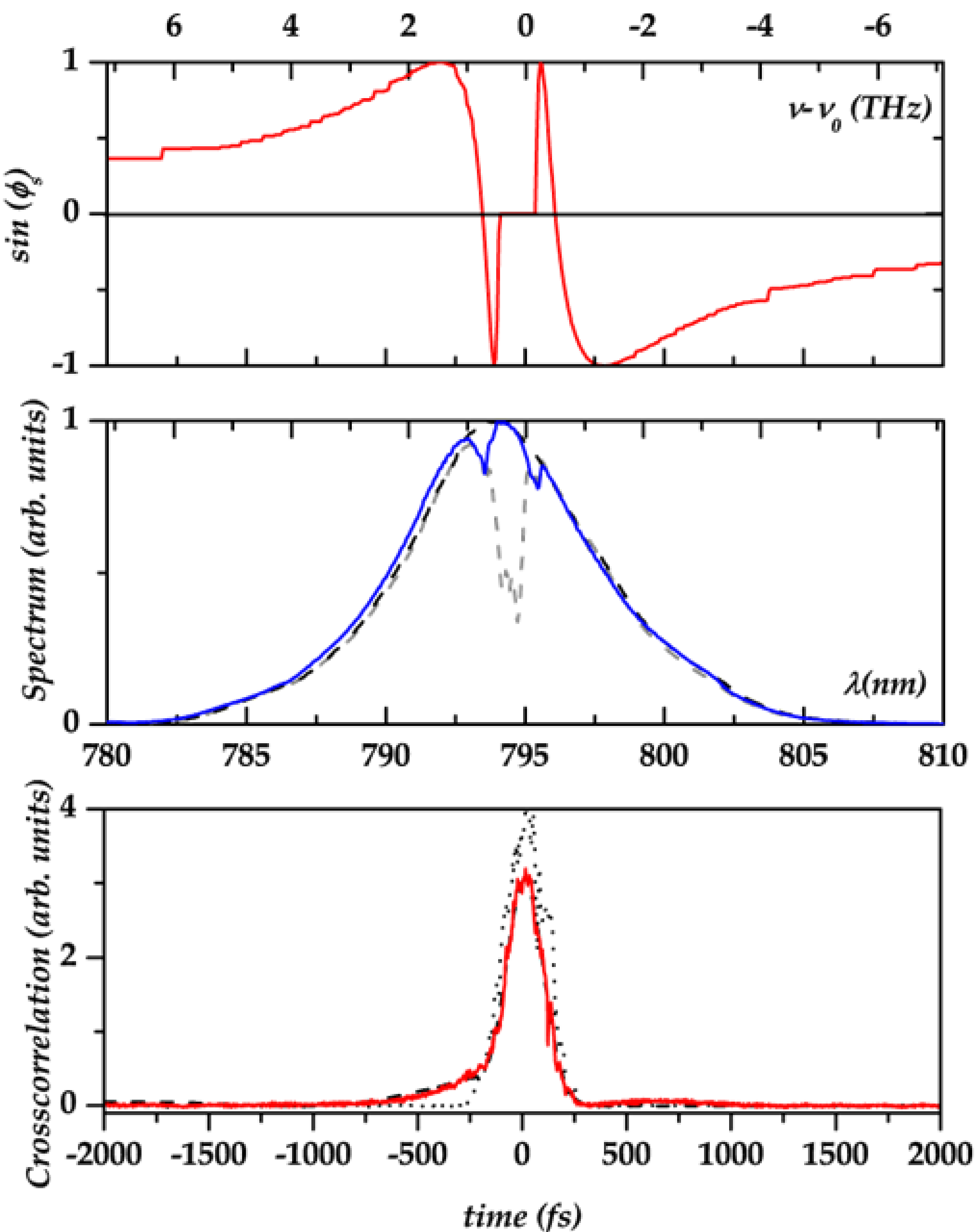
⇒ Spectral hole around λ !

Compensation with a pulse shaper

- Asymptotic part well reproduced
- Unable to reproduce exact behaviour near the resonance
- Spectrum intensity is afffected

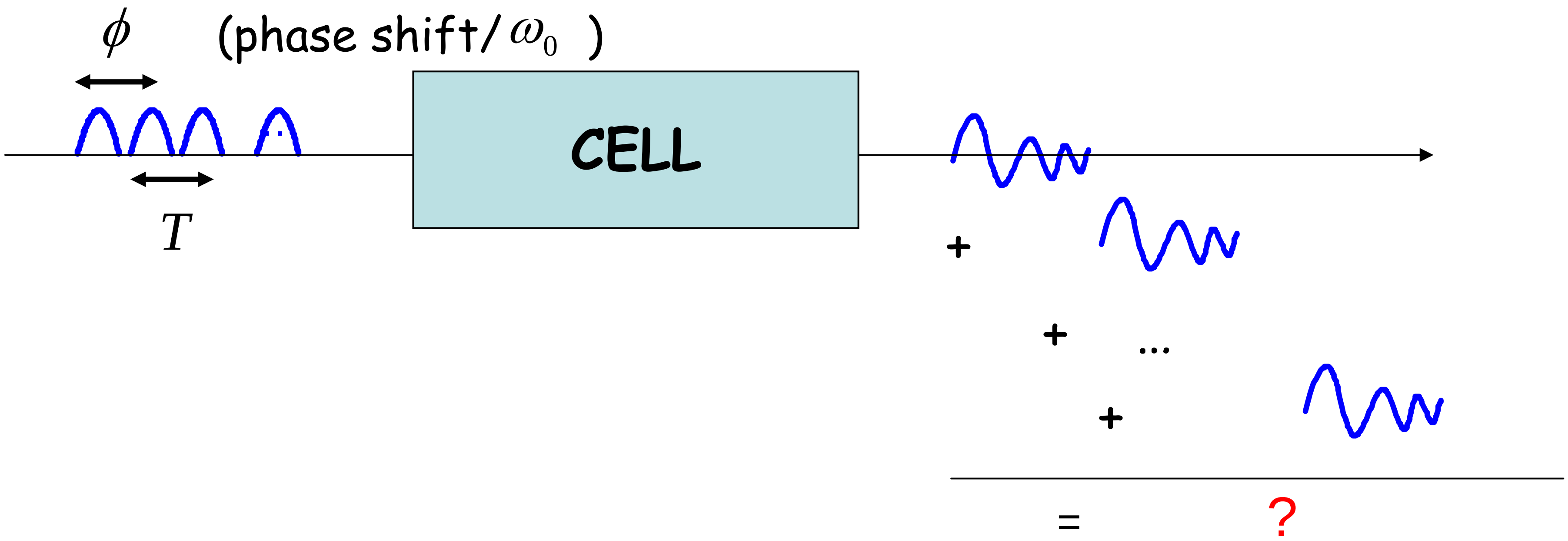


Flat Phase



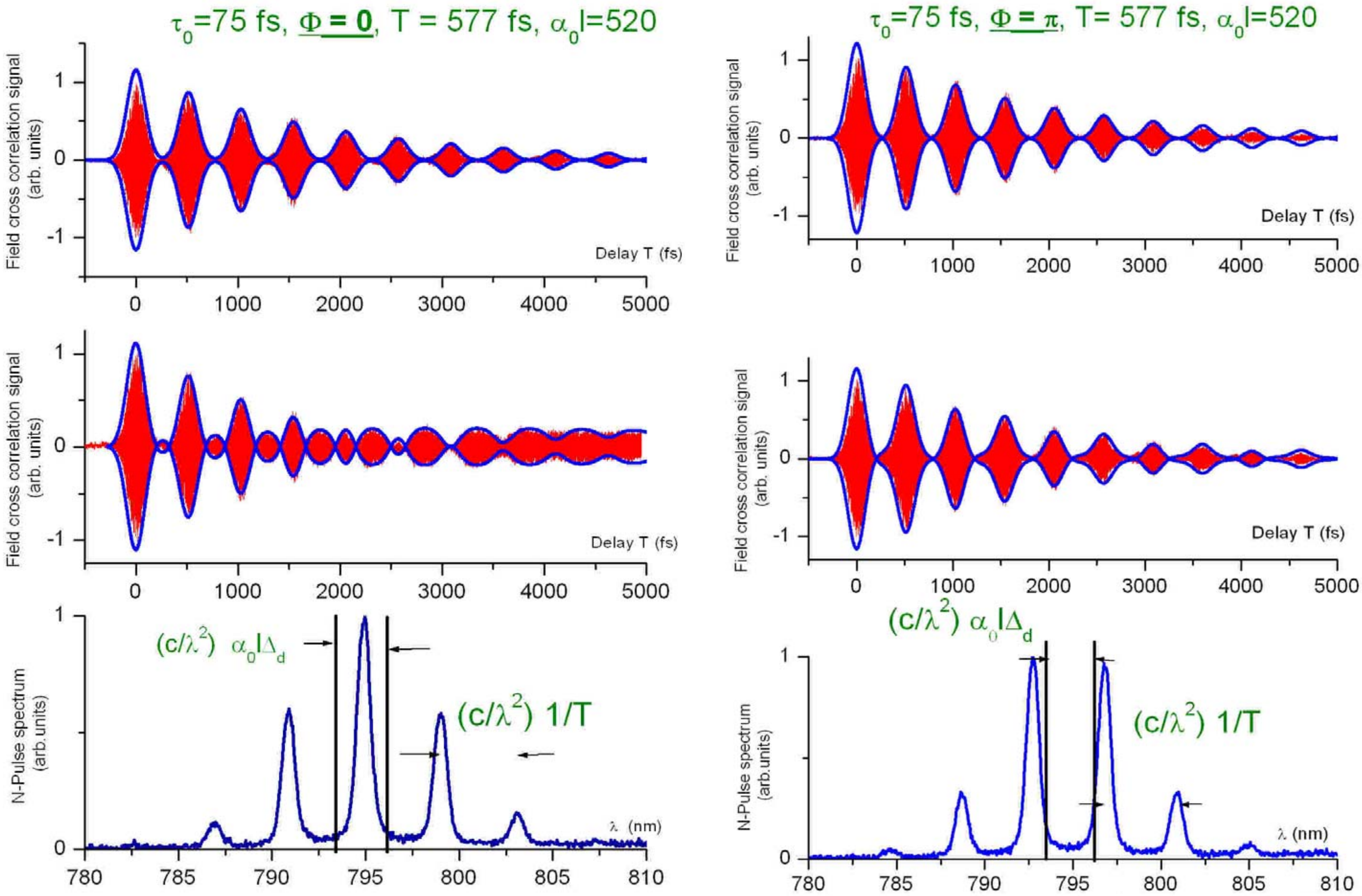
6
4
2
0
-2
-4
-6
1
0
-1
ν-ν_0 (THz)
$sin(\phi_s)$
Spectrum (arb. units)
1
0
λ(nm)
780
785
790
795
800
805
810
Crosscorrelation (arb. units)
4
2
0
-2000
-1500
-1000
-500
0
500
1000
1500
2000
time (fs)

3. Case of an ultrashort pulse train

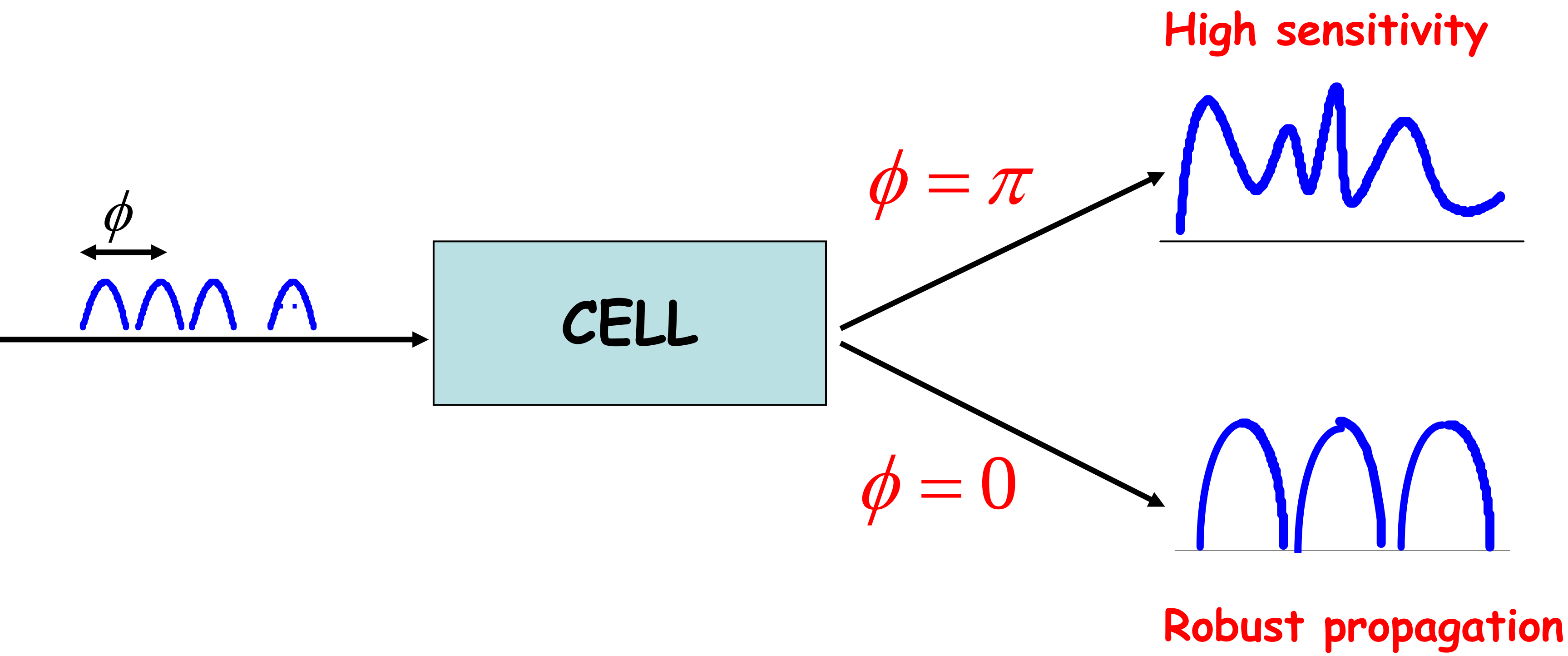


- **Independent pulses** : Intensity superposition.
- **Mutually coherent pulses**: Field superposition .

→ Depends on both ϕ and T

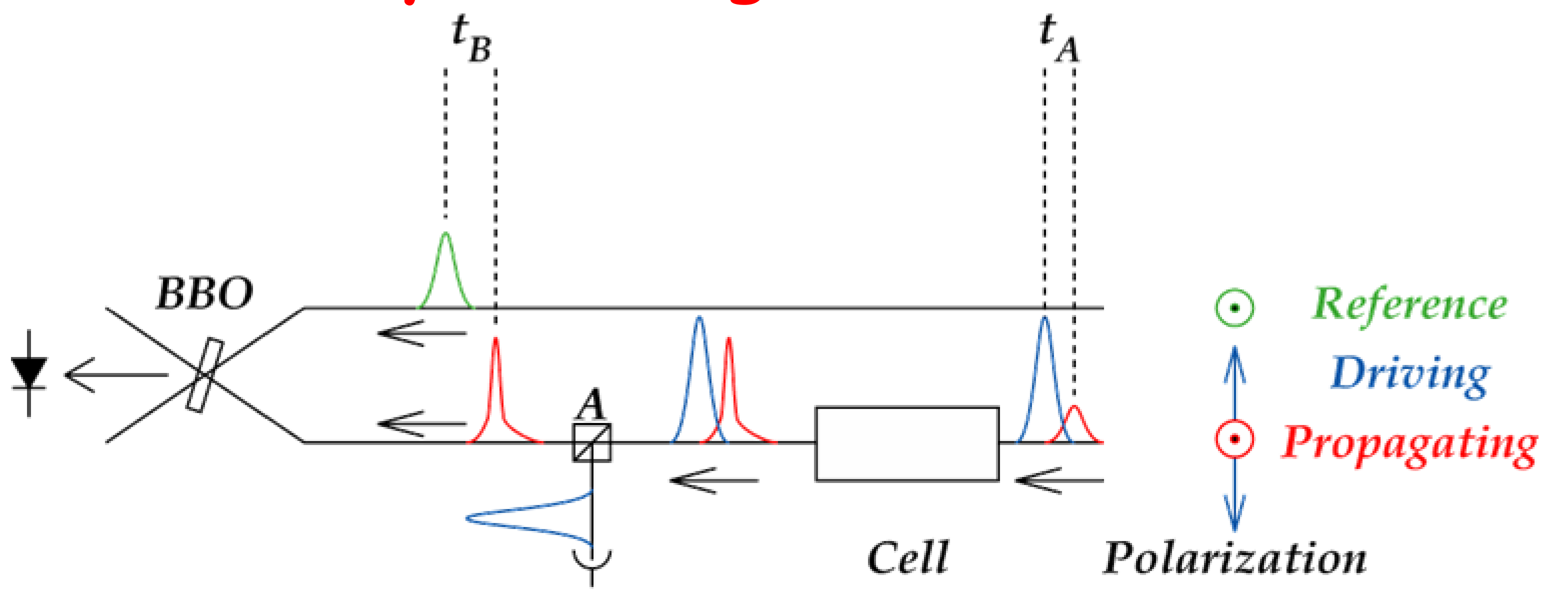


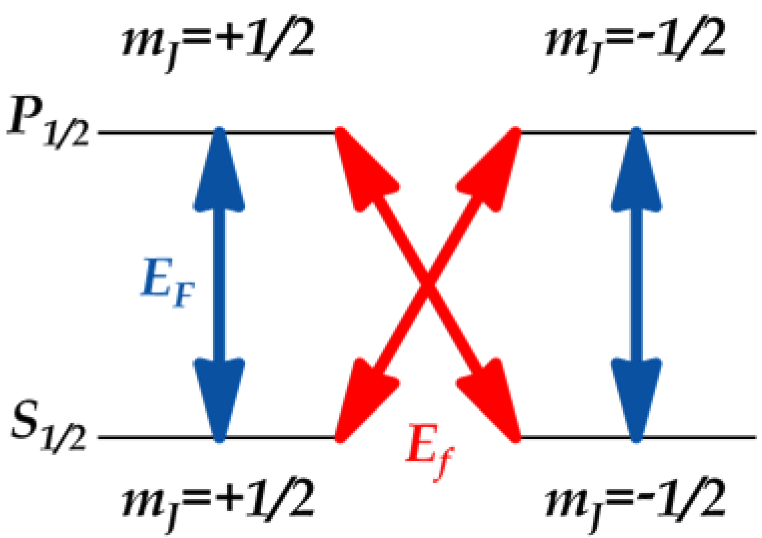
Negligeable if $\alpha_0 l \Delta_d < T^{-1}$; $\alpha_0 l)_{max} \sim \tau_0^{-1}/\Delta_d$



PHASE CONTROL OF DISPERSION EFFECTS

4. Propagation in an atomic system driven by a strong field



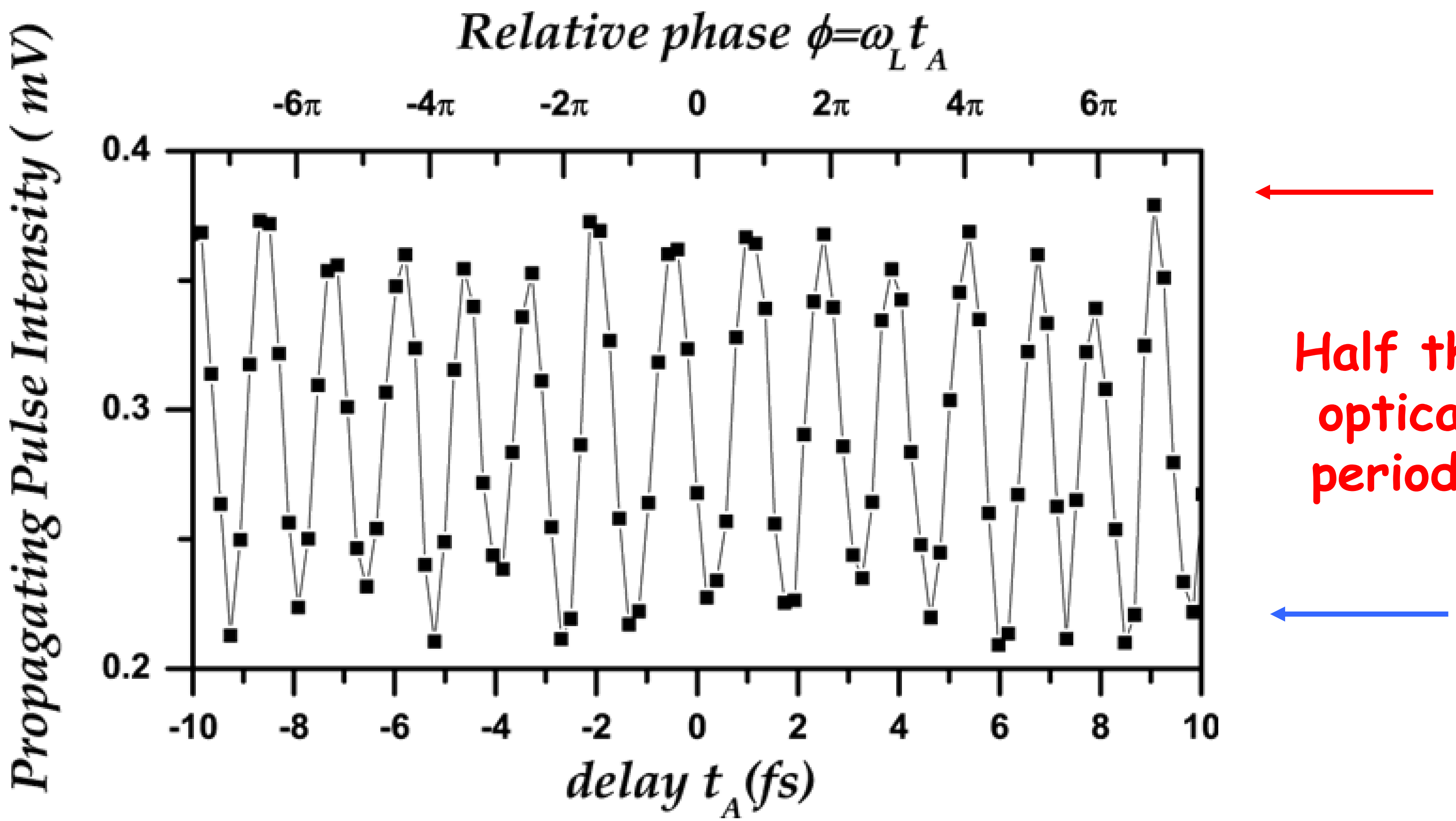


Modifications due to the Strong Field (effect of Relative Phase and Intensity)

On the energy and temporal Profile of the propagating pulse

Rb atom $4s\, S_{1/2} \rightarrow 4p\, P_{1/2}$

Coherent Control of the Gain

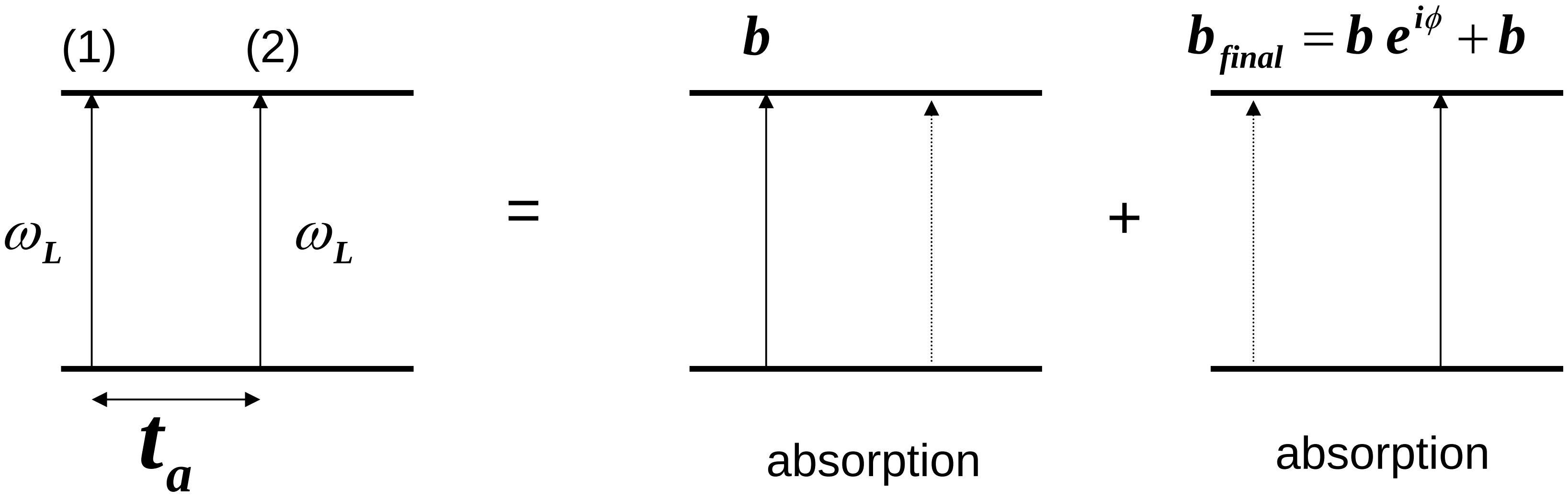


Half the optical period !!

- Crossed polarisation !
- Interference at $2\omega_L$ in one photon transition!!!!

Interpretation

1- « Ordinary » interference in one photon transition (Temporal Ramsey fringes)

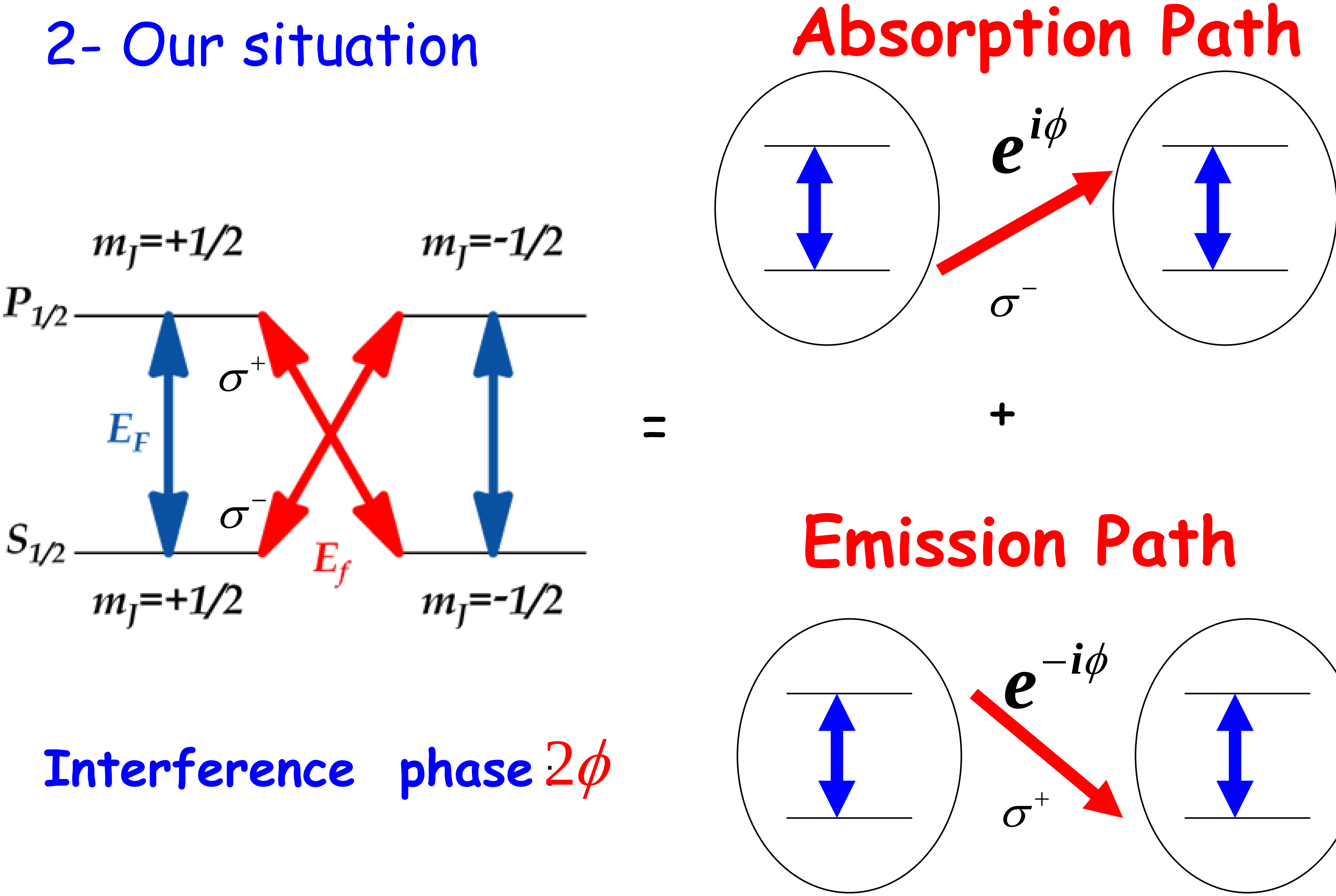


Looking at the population the excited state:

$$n_f = 4n \cos^2 \phi / 2$$

Interference between two absorption paths phase-shifted by $\phi = \omega_L\, t_a$

2- Our situation

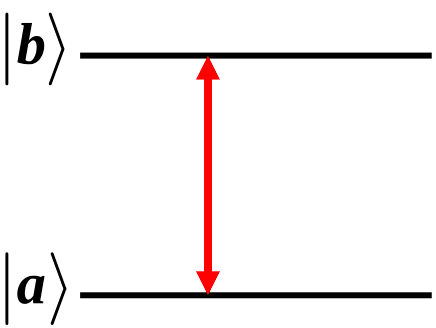


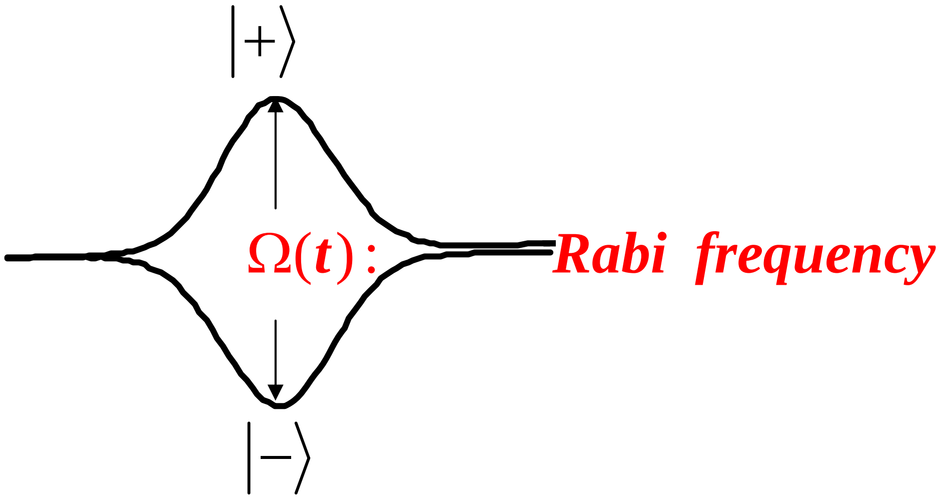
Interference phase 2ϕ

1- Interf. between absorp. and emis. paths connecting two linear superp. of states
2- Interference phase 2ϕ , $\phi = \omega_L \, t_a$ the phase with respect to the strong field.
3- The two paths are « synchronous » (phase shifted but not delayed!)

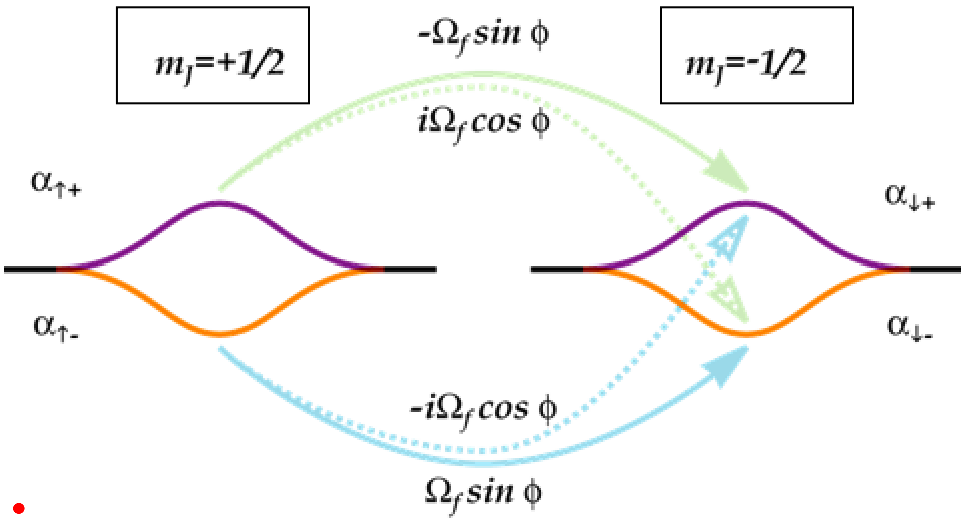
Dressed state analysis

$$\left(|a\rangle, |b\rangle\right) \quad \longleftrightarrow \quad |\pm\rangle = \frac{\mp|a\rangle + e^{-i\omega_0 t}|b\rangle}{\sqrt{2}}$$





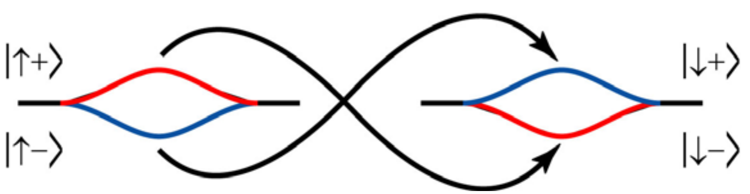
Action of the weak field

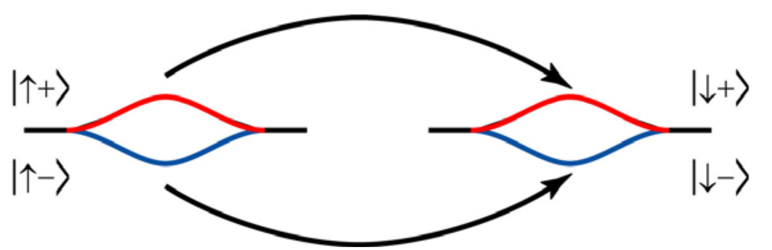


Two situations

$$\phi = 0$$

$$\phi = \frac{\pi}{2}$$

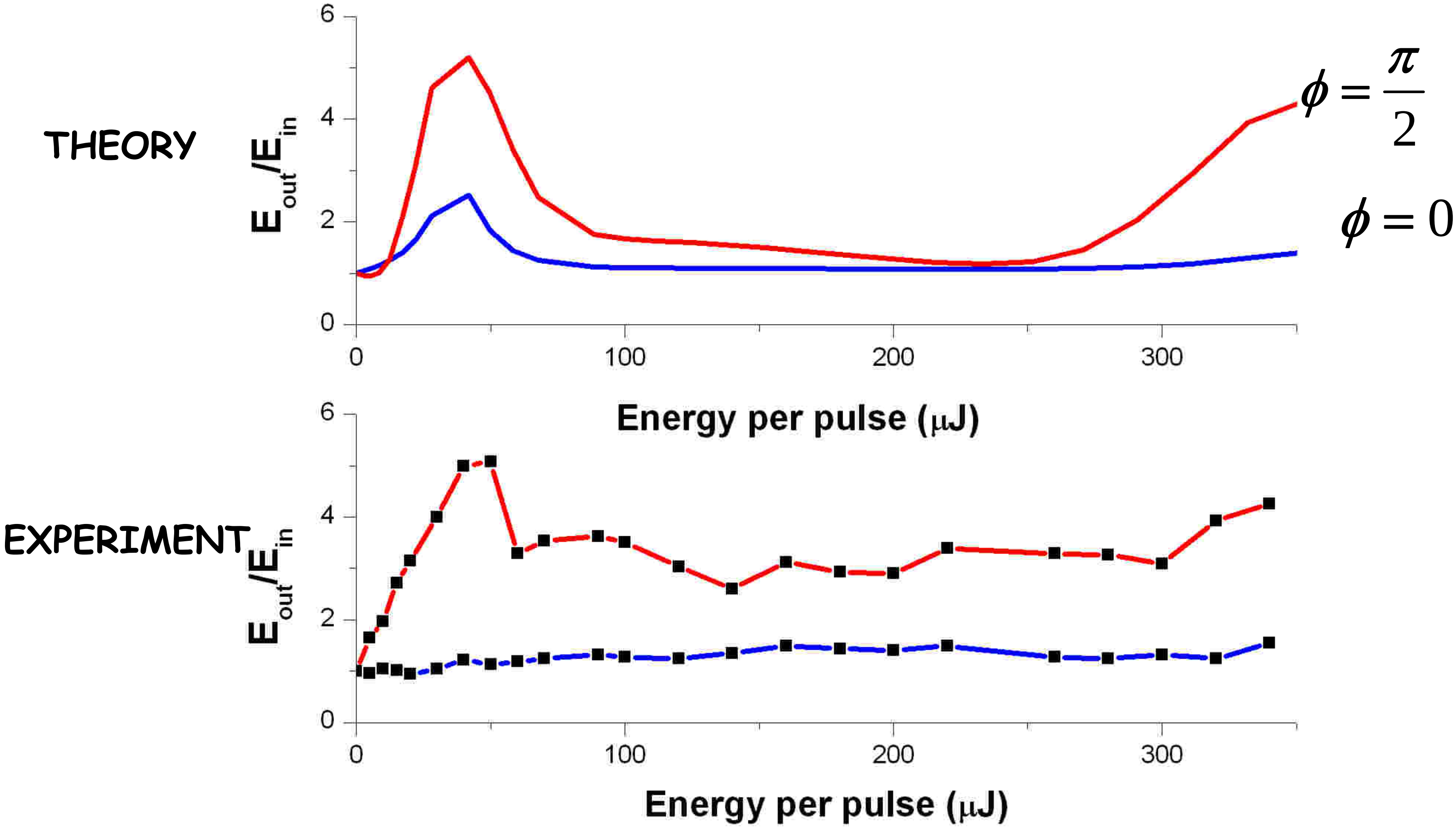




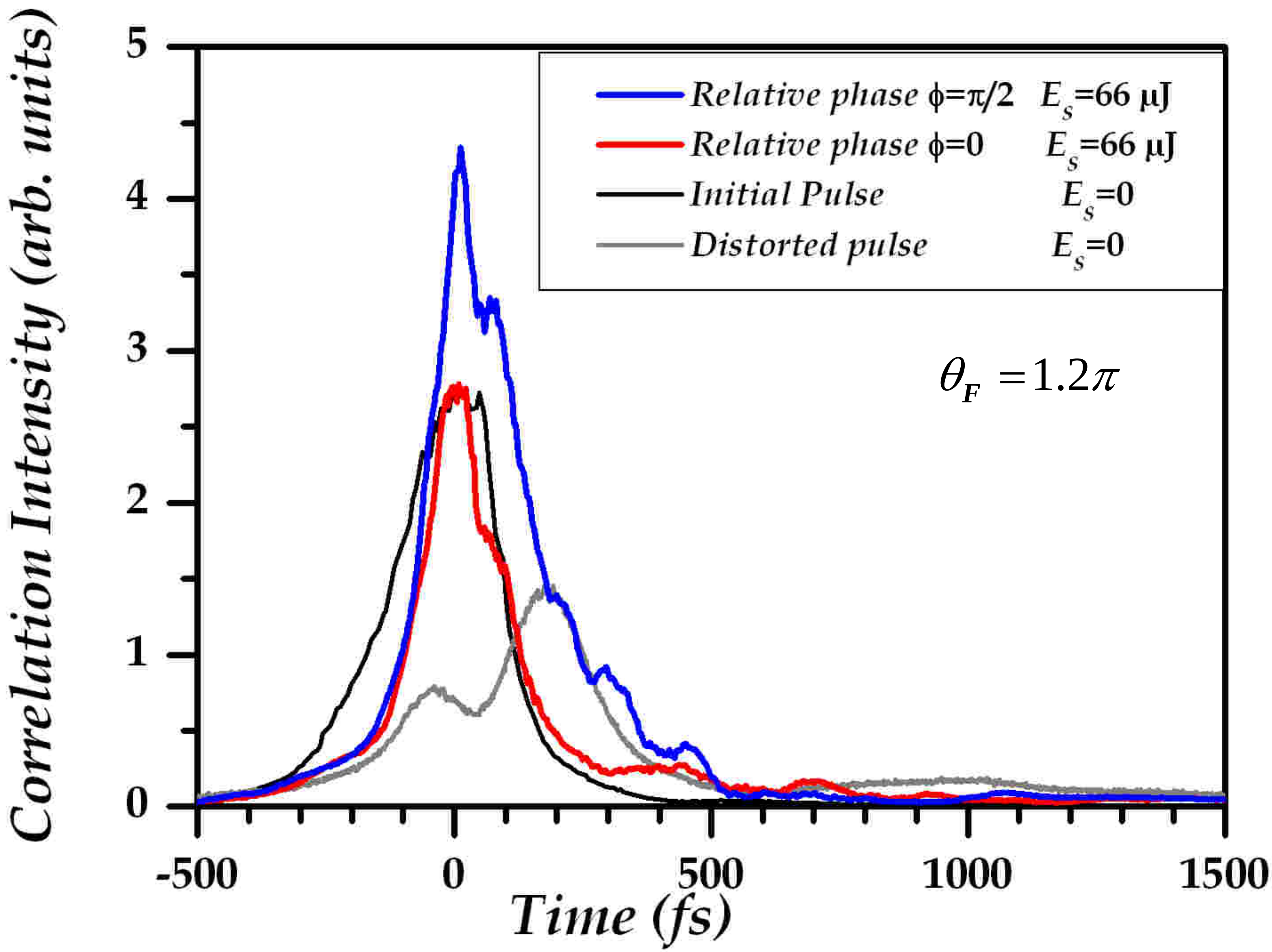
Transparency window
Non-resonant

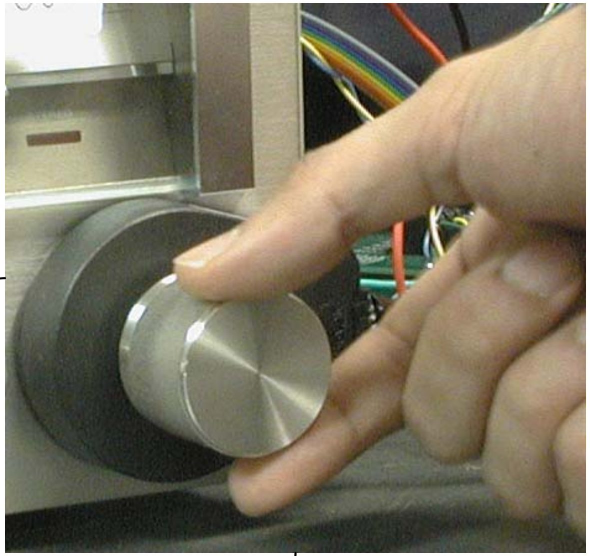
Absorption/amplification
Resonant

Dependence vs Strong Field energy at Zero Delay



Control of the Shape





Control Field Strength θ_F

Energy

Shape

Phase ϕ

5. Towards « active » pulse shaping

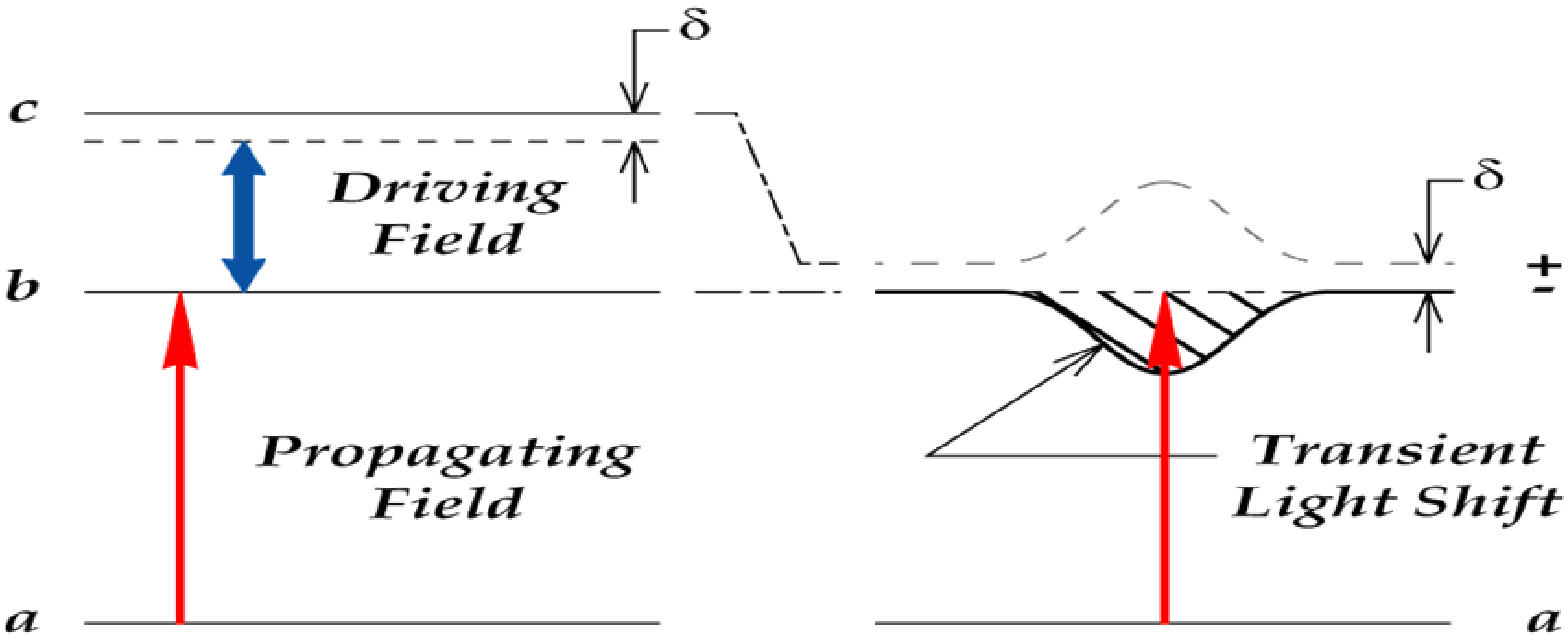
Classical devices (pulse shaper)

Passive : no amplification
no creation of spectral components

Strongly driven system

Active: create new frequencies
(light-shift)

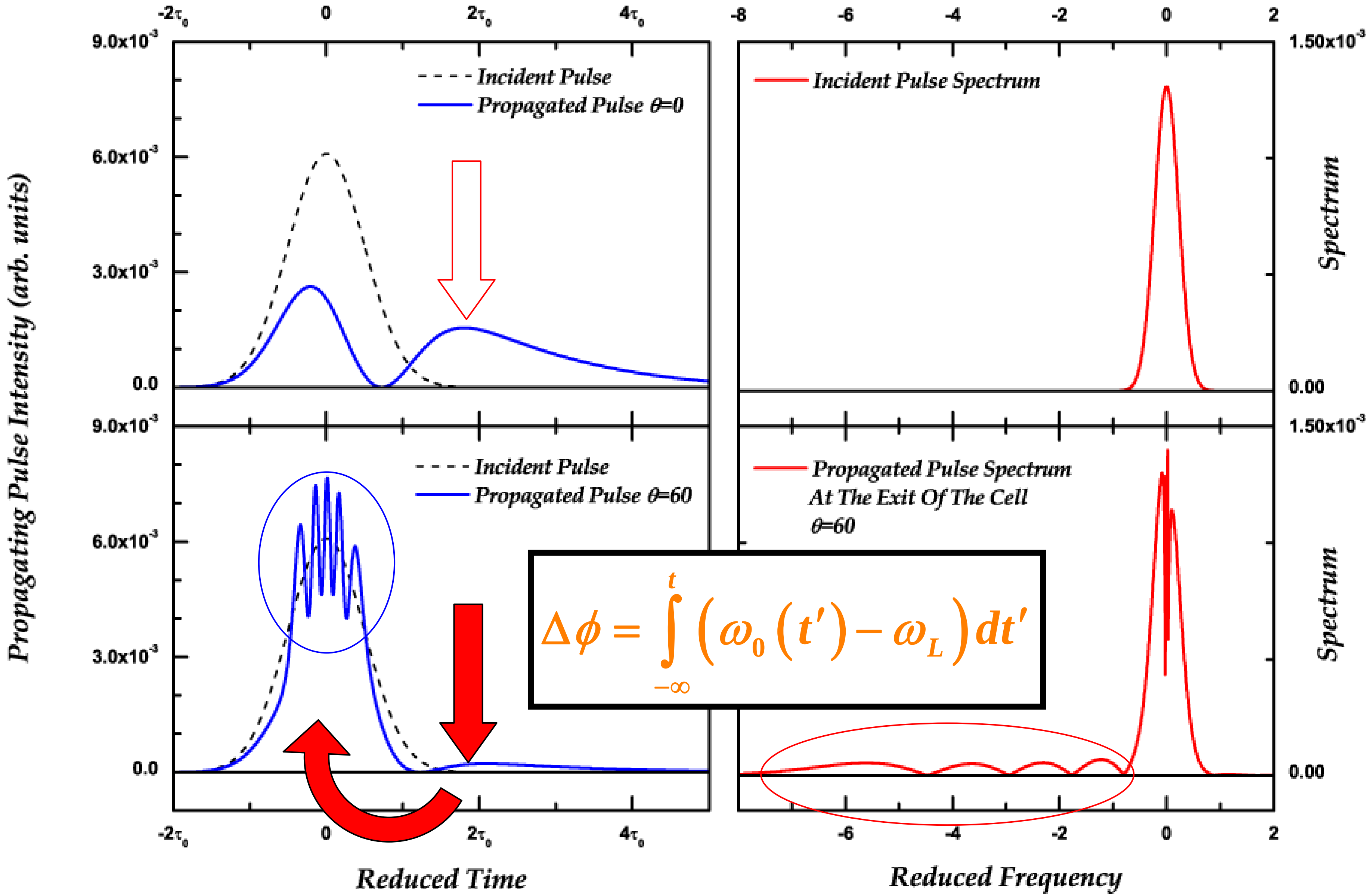
Transient Light Shift in a 3 level Ξ system



$$E_{out} = E_{in} + E_{rad}$$

$$\propto e^{-i\omega_L t} + \propto e^{-\int_{-\infty}^{t} \omega_- dt'}$$

Fixed Laser Frequency + Varying Dipole Frequency



Propagating Pulse Intensity (arb. units)
Incident Pulse
Propagated Pulse θ=0
Incident Pulse
Propagated Pulse θ=60
Incident Pulse Spectrum
Propagated Pulse Spectrum
At The Exit Of The Cell
θ=60
Spectrum
Spectrum
$\Delta\phi = \int_{-\infty}^{t} \left(\omega_0\left(t'\right) - \omega_L\right) dt'$
Reduced Time
Reduced Frequency

Varying Dipole Frequency

+

Fixed Laser Frequency

$$\Delta\phi = \int_{-\infty}^{t} \left(\omega_0\left(t'\right) - \omega_L\right) dt'$$

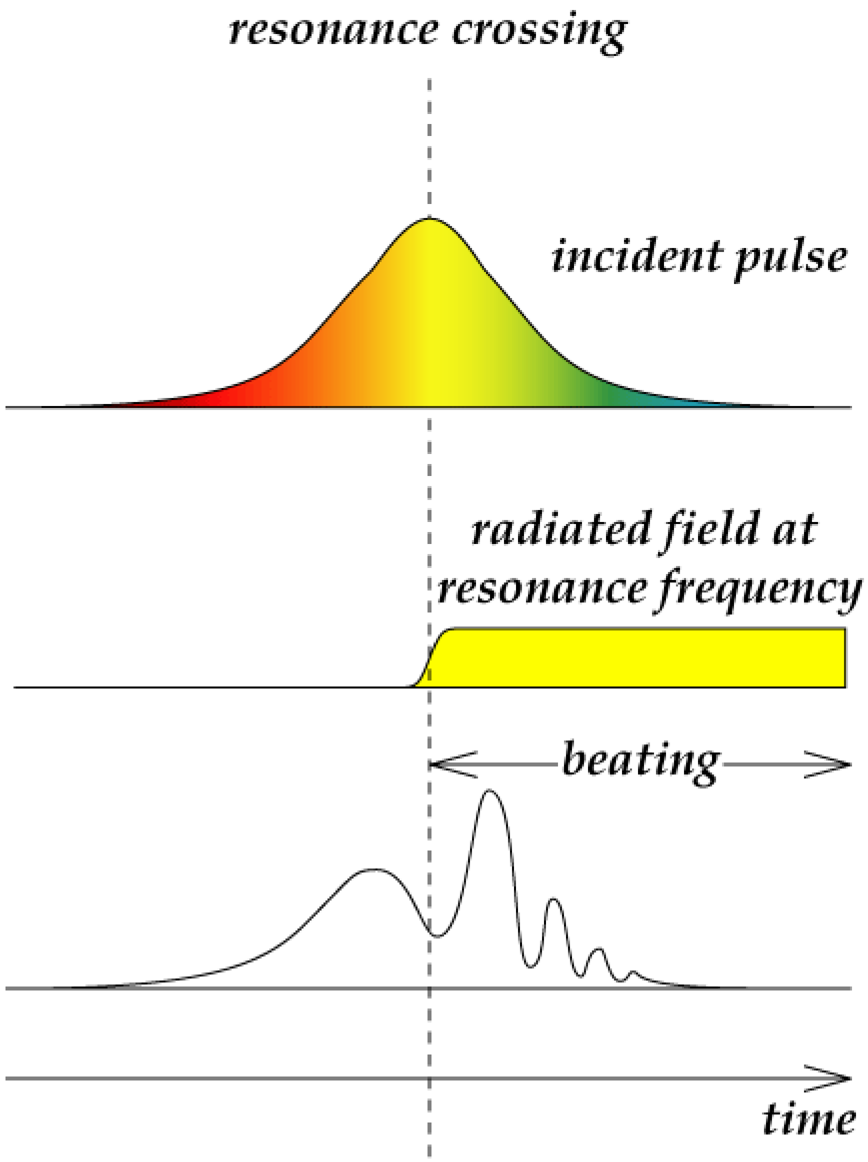
Fixed Dipole Frequency

+

Varying laser Frequency

$$\Delta\phi = \int_{-\infty}^{t} \left(\omega_0 - \omega_L\left(t'\right)\right) dt'$$

Chirped pulse propagation: principle

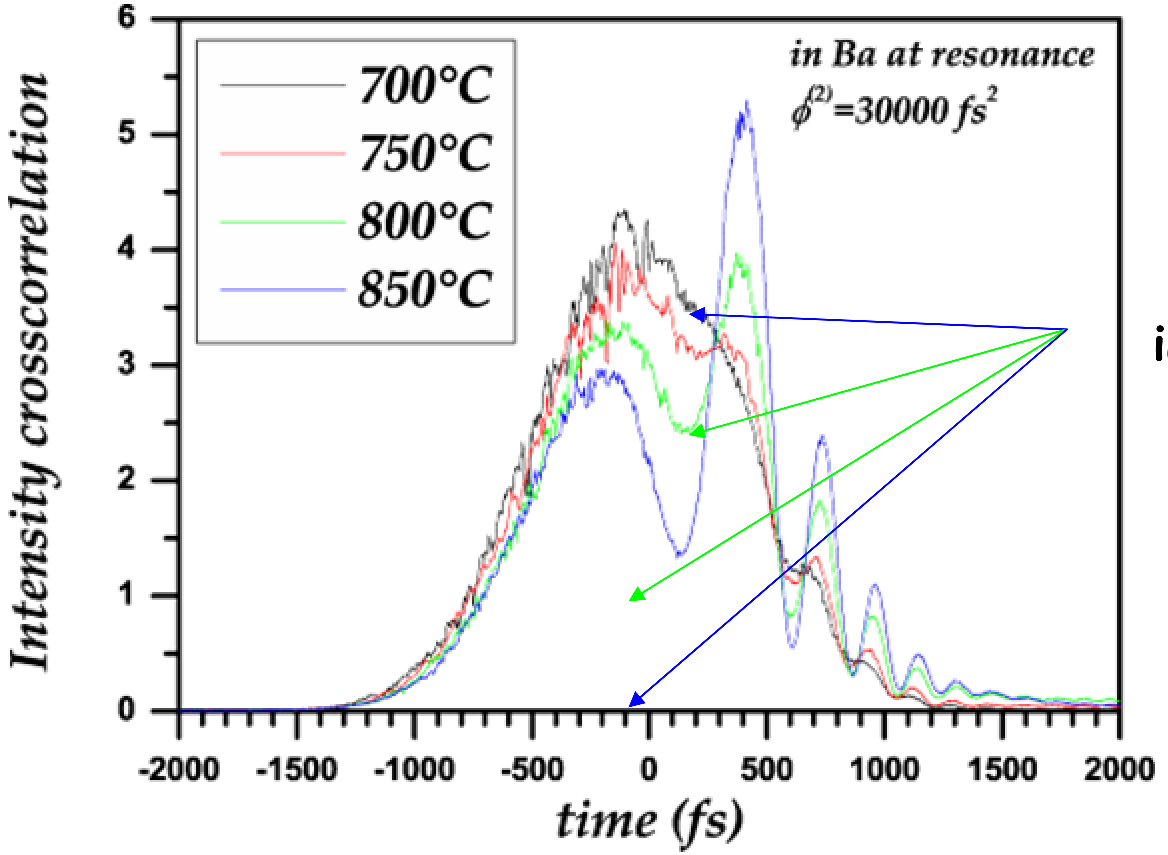


- **<u>Self-induced heterodyne field</u>**
- **Mapping of the incident field phase on the intensity profile**

$$\Delta\phi = \int_{-\infty}^{t} \left(\omega_0 - \omega_L(t')\right) dt'$$

- **<u>Direct basic temporal shaping</u>**

Chirped pulse propagation: experiment



Depth of modulation increases with the density

Conclusion

- Atomic system at equilibrium:

  Compensation of Dispersion for a weak pulse and a pulse train

- Strongly driven atomic system :

  2ω Oscillations on one photon transition

  - **Coherent Control of Energy**
  - **Coherent Control of the pulse Shape and possibility of active pulse shaping**