Coherent Control in Small Systems : Spin Dynamics, Cold Molecules....

IRSAMC
Laboratoire Collisions Agrégats Réactivité
University of Toulouse-CNRS (France)
Béatrice Chatel

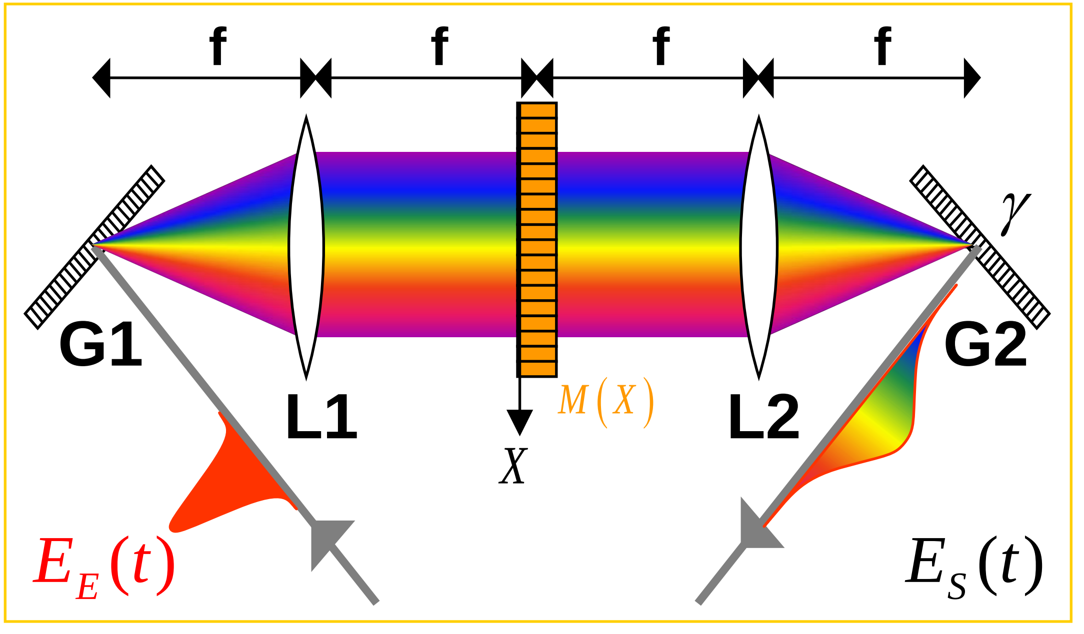
B. Girard, S. Weber, M. Barthélémy, C. Handschin

Coherent control :

- To take advantage of several quantum paths during the interaction.
- To play with interferences…
- How? By controlling laser parameters (energy, wavelength, spectral phases, spectral amplitude, polarisation…)

Our tools:
ultrashort pulses and pulse shaper

$$E(t) = |E(t)| e^{i\varphi(t)} \quad \underset{\mathcal{F}^{-1}}{\overset{\mathcal{F}}{\rightleftarrows}} \quad \tilde{E}(\omega) = |\tilde{E}(\omega)| e^{i\phi(\omega)}$$



Linear passive Filter

$$\tilde{E}_S(\omega) = H(\omega)\tilde{E}_E(\omega)$$

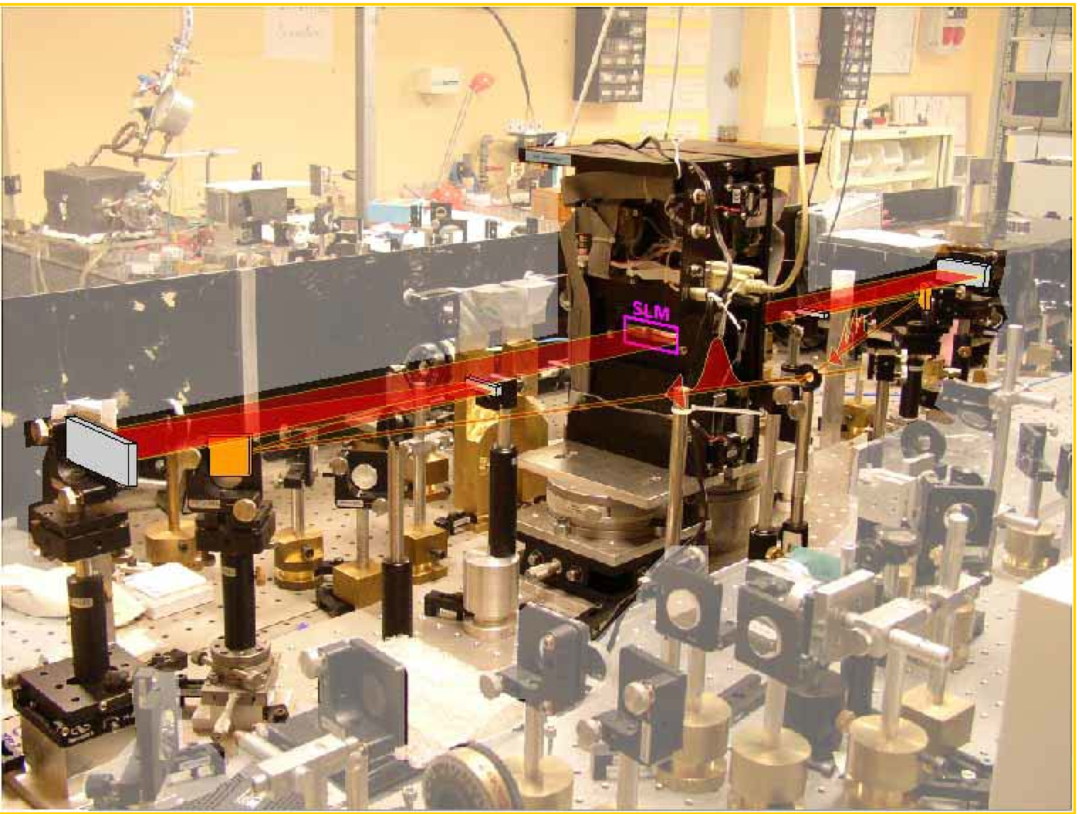
with

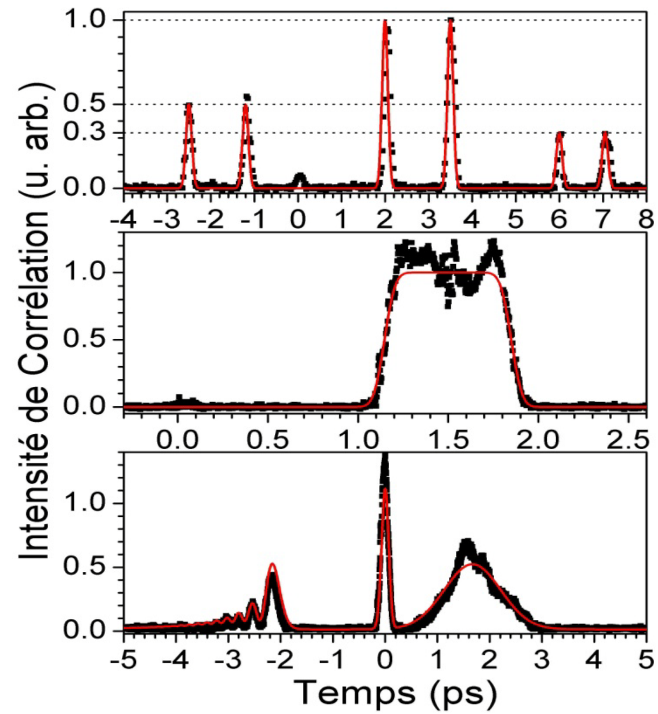
$$H(\omega) = A(\omega)e^{i\phi(\omega)}$$

Weiner, A.M., *RSI*, **71**, (5), **2000**

$$\tilde{E}_S(\omega) = M(f\gamma.\omega)\tilde{E}_E(\omega)$$

Our high resolution pulse shaper





- Phase/Amplitude control over 640 pixels.
- shaping window of 35 ps.
- 0.06 nm/pixel
- high amplitude dynamic (30 dB).
- 75 % power transmission.

A. Monmayrant, B. Chatel. *"A new phase and amplitude **H**igh **R**esolution **P**ulse **S**haper."* ***Rev. Sci. Inst.*** **75,** 2668 (2004)

Three examples

- Coherent control to cool molecules
- Coherent control to manipulate spin precession in atoms
- Coherent transient revisited : Shape the pump and the probe

Cold molecules, Why ?

Precise measurement

Fundamental test (e- dipole, chirality, constant variation)

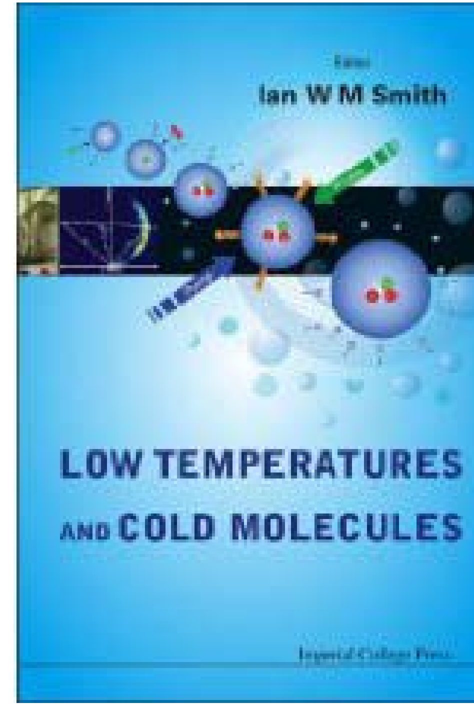
Quantum information, computation, logic…

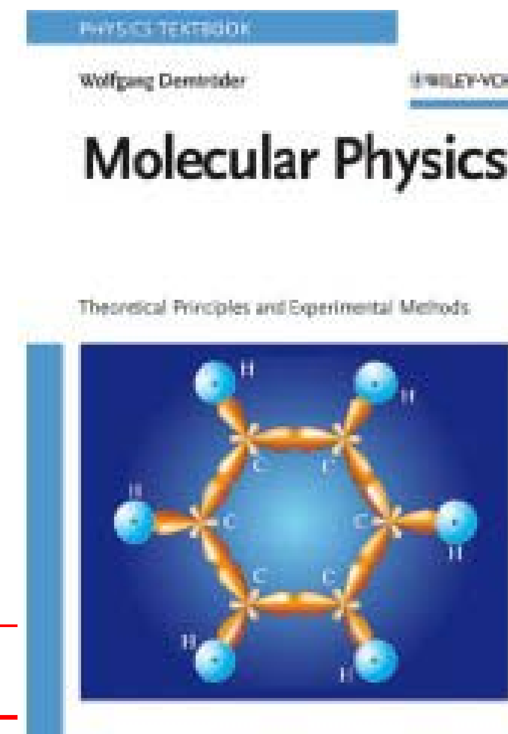
Quantum properties (dipole), BEC,BCS

Control of (Reactive) collisions: quantum chemistry
Photochemistry (photoassociation),
Superchemistry (Bose-Enhancement)

1. J. Doyle, et. Al *Eur. Phys. J. D* **31**, 149 (2004).
2. R. V. Krems, Phys. Chem. Chem. Phys.10, 4079 (2008).
3. J. Hutson, *Int. Rev. Phys. Chem*. 25, 497 (2006).
3. O. Dulieu, M. Raoult, E. Tiemann, *J. Phys. B* **39** (2006).

Need ultra-cold molecules (T~0K) and in v=J=0

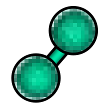




To cool molecules

From molecules

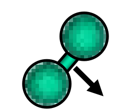
- Cryogenic cooling
- Slowing of supersonic beam
- Velocity filtering



Translation
hot
+
Vibration
cold

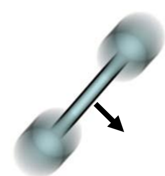
GOAL

Translation
cold
+
Vibration
cold



From atoms

- External field (Feshbach) @ 1μK, 10^{12} at/cm^3
- Collision
- Photon-association @ 100μK, 10^{10} at/cm^3

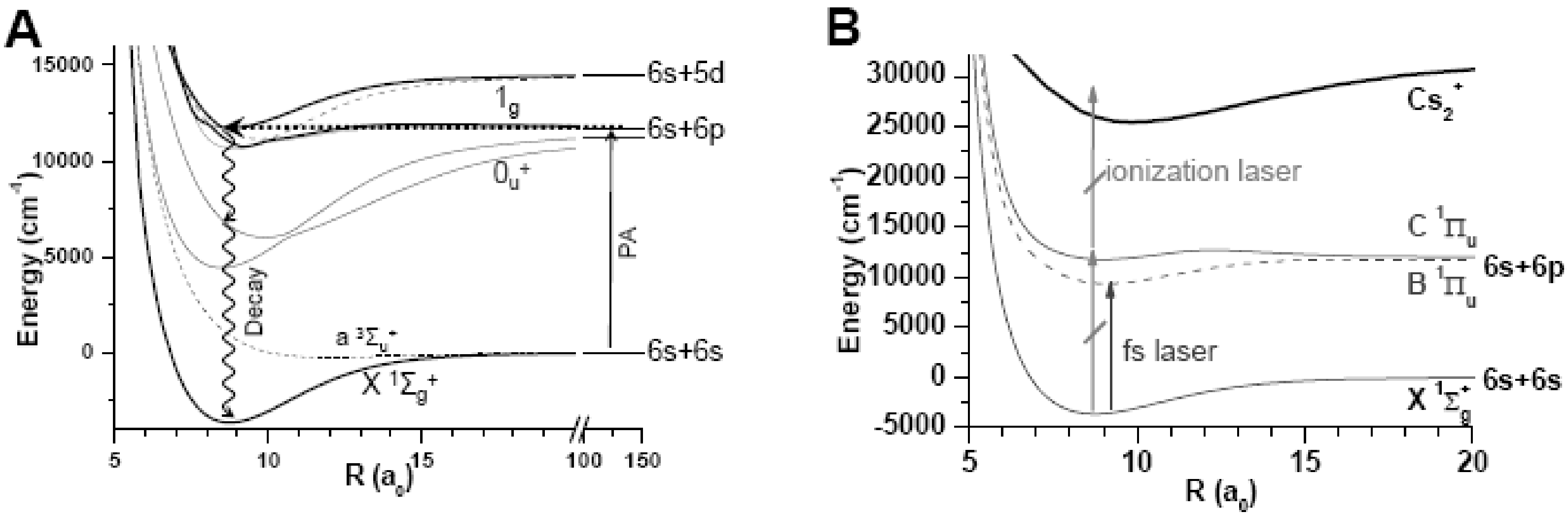


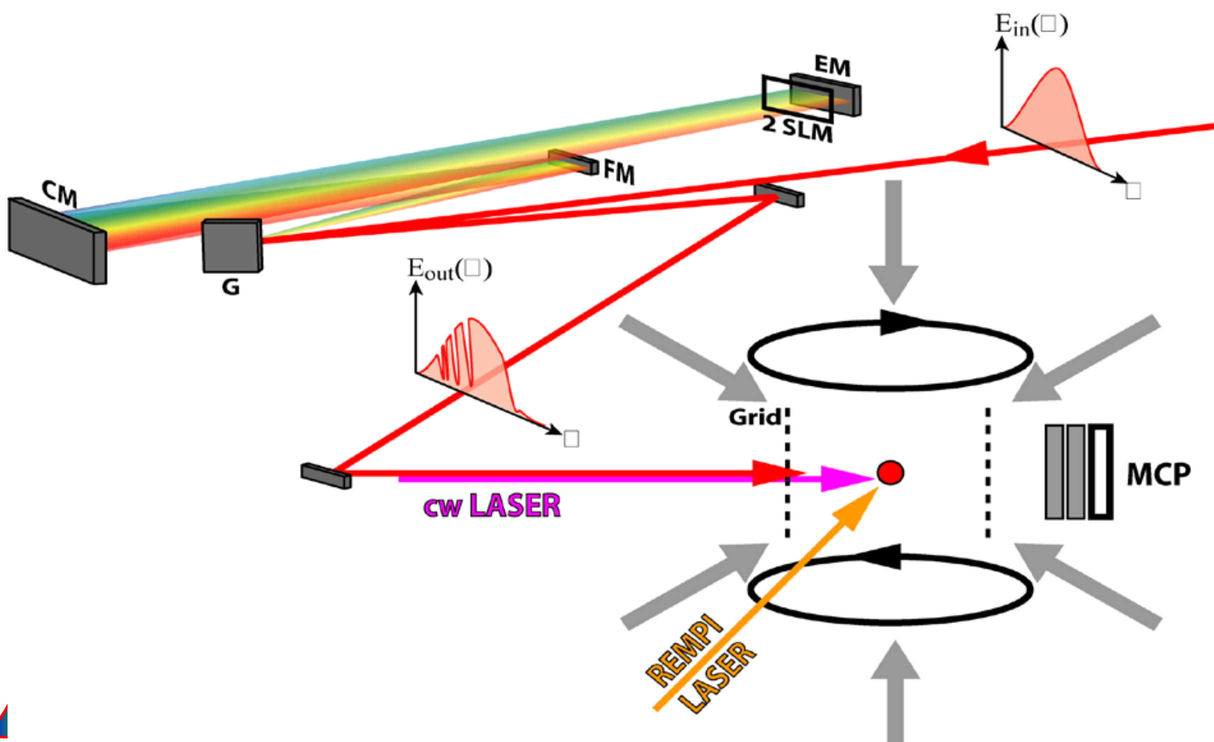
Translation
cold
+
Vibration
Hot (not v=0)

Two approaches to cool molecules based on photo-association

- Use coherent control scheme as pump-dump : preliminary works in coll with I. Walmsley (Oxford)
    - arXiv:0904.0244 (PRA-2009)
    - PRL96, 173002 (2006).
- Optical pumping in coll with D. Comparat/P. Pillet (LAC-France) :M.Viteau,A. Fioretti….
    - Incoherent approach
        - Science 321, 232 (2008)
        - Phys. Rev. A 79, 021402(R) (2009)
        - New J. Phys. 11 (2009) 055037.

Incoherent approach





Makes cold molecules : Long Range Molecules (high v, but low J).

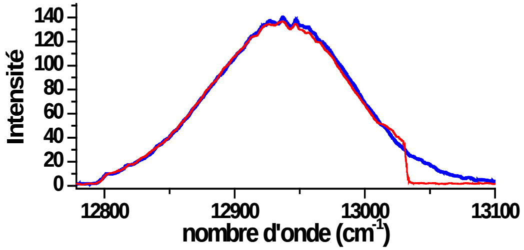
Several vibrational states populated

Then optical pumping to cool the vibration

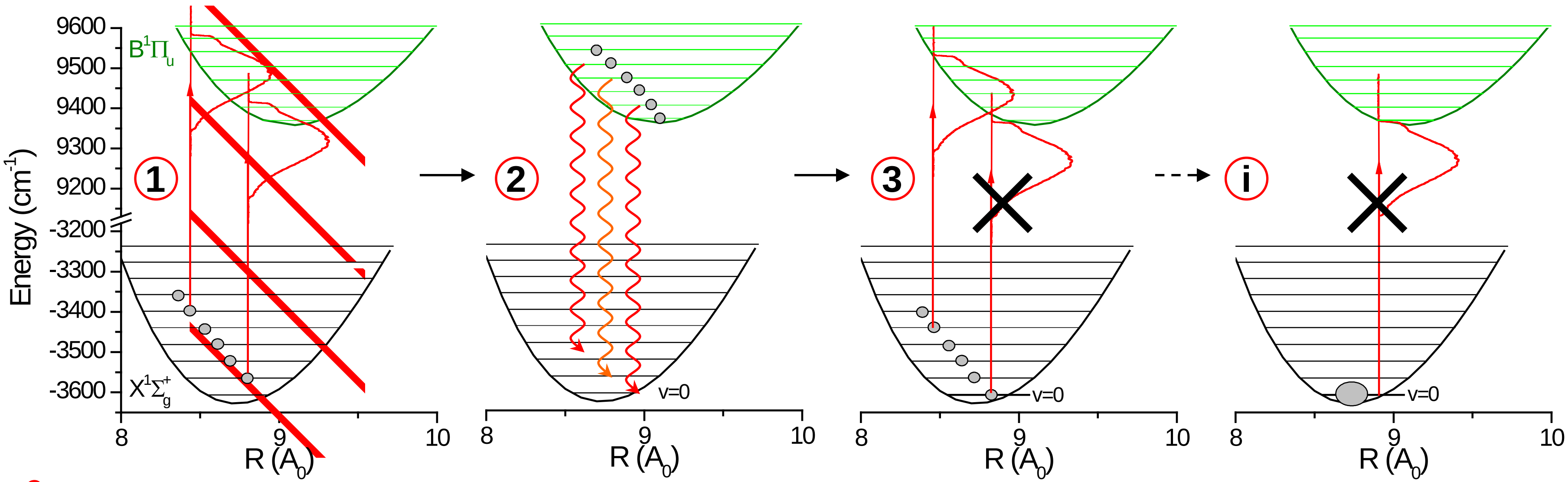
Optical pumping and vibrational cooling

Viteau, Comparat, Pillet et al

Science 321, 232 (2008)



To use the laser to excite all populated vibrational levels but frequency-limited in such a way to eliminate transitions from v = 0 level, in which molecules accumulate (creation of a dark state)



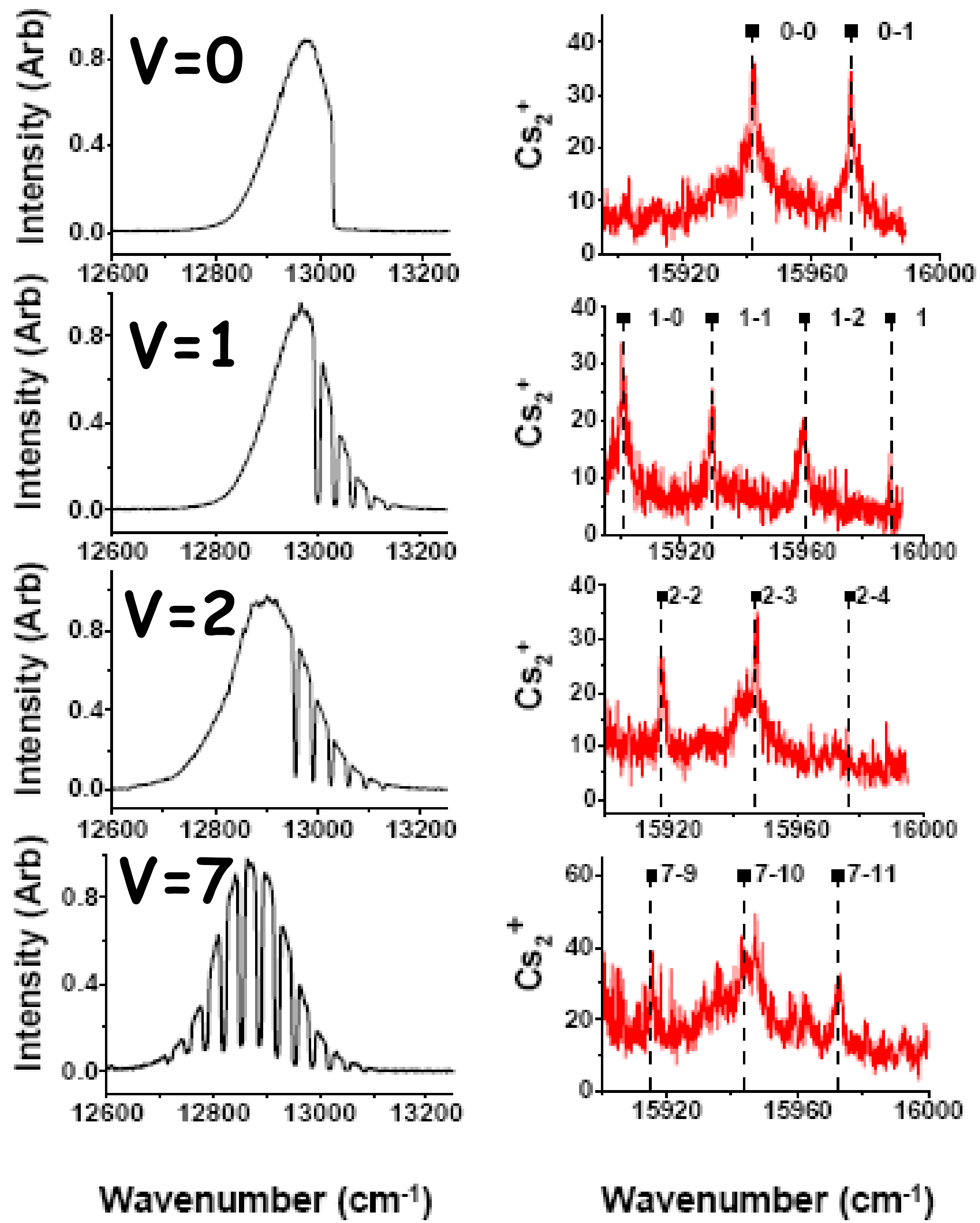
Choosing the dark state

REMPI ion spectrum

a)

To extend this incoherent optical pumping in order to accumulate molecules into other single selected vibrational level than the sole v = 0 one.

c)

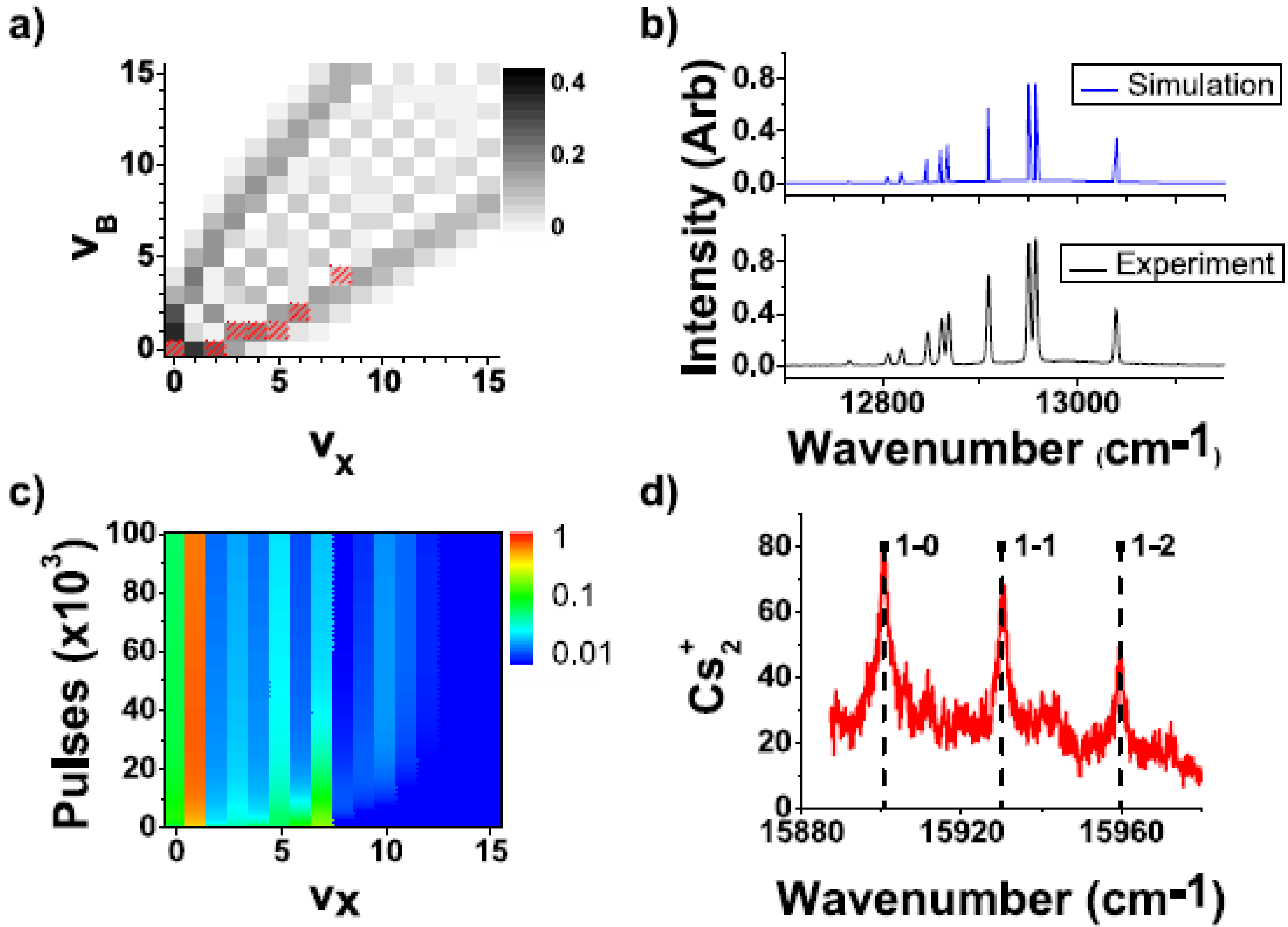


Very fast (<100 μs),10^4 pulses
Only few cycles to transfer 60% of the molecules
→ Almost no heating

Sofikitis, Chatel *et al.*

New J. Phys. 11 (2009) 055037.

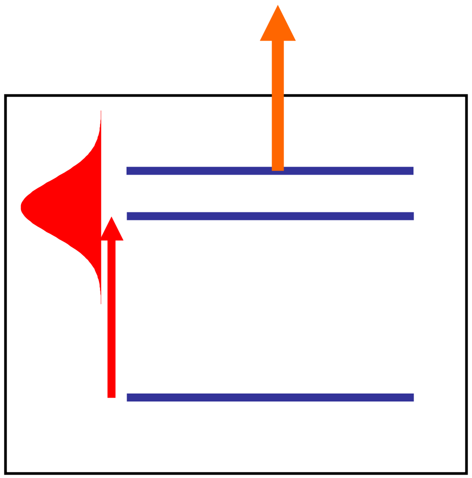
Using a comb to favour high FC factor



Three examples

- Coherent control to cool molecules
- **Coherent control to manipulate spin precession in atoms**
- Coherent transient revisited : Shape the pump and the probe

Quantum control using pathways interferences



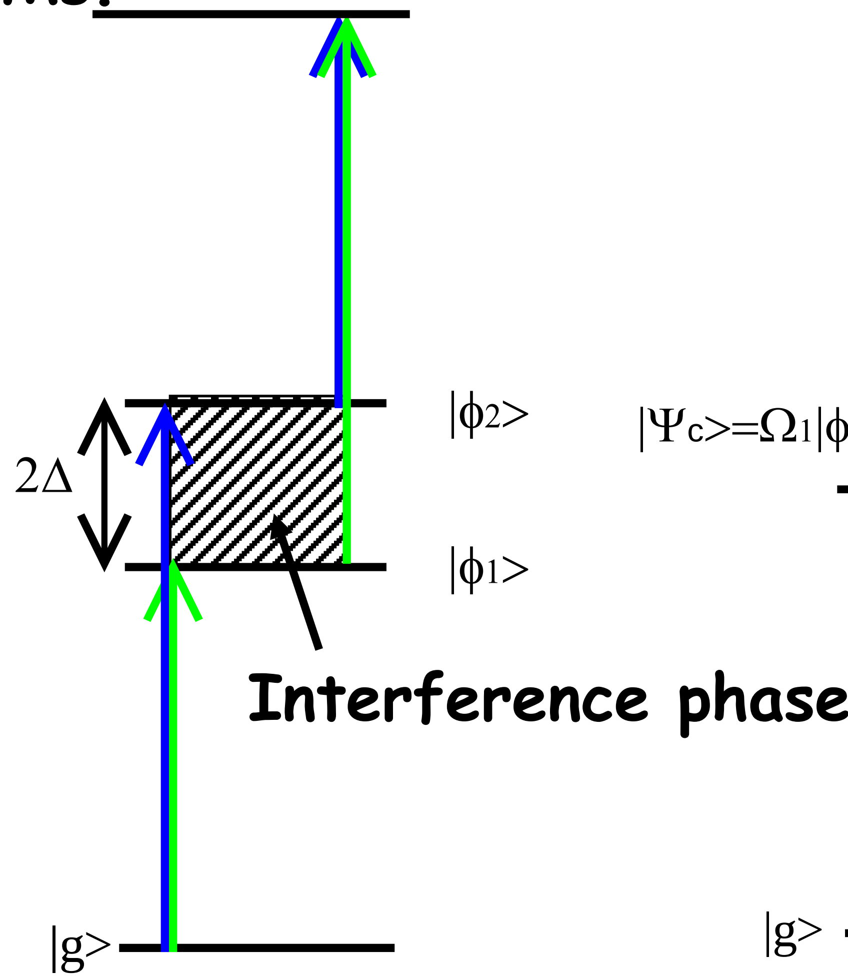
Excitation of a superposition of two states

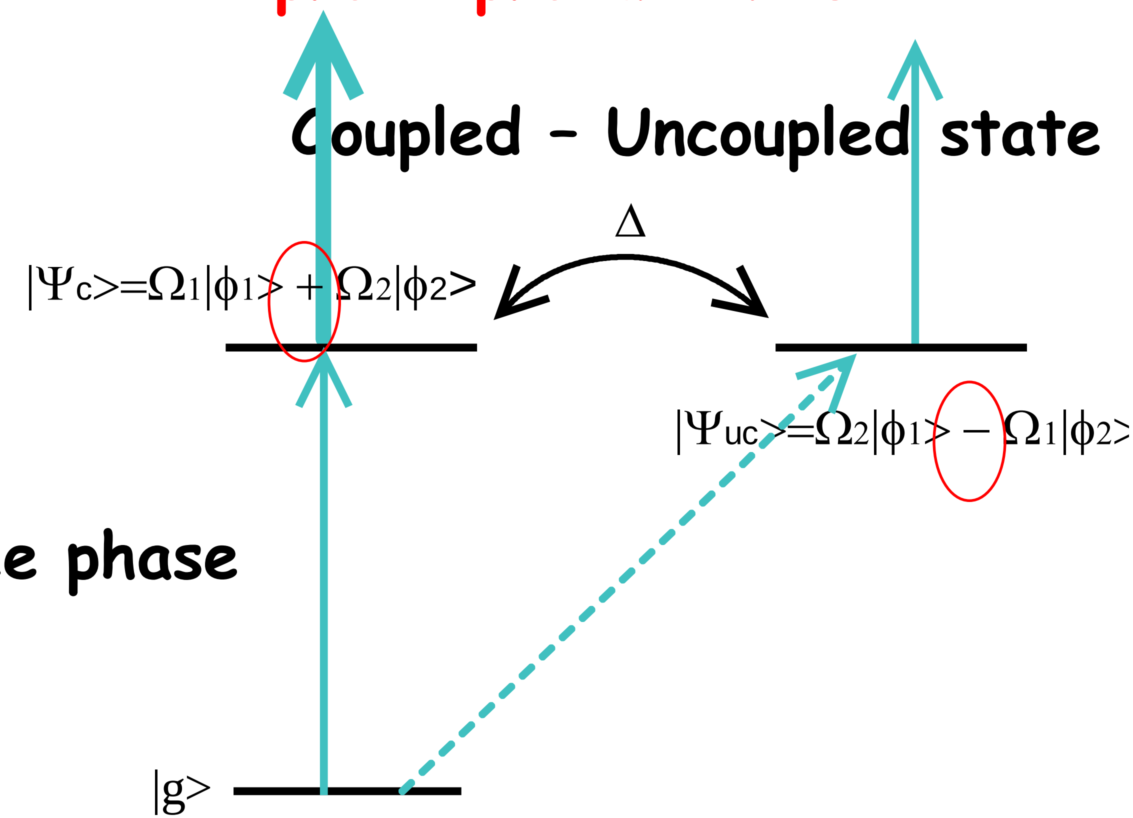
Here we consider a pump probe experiment which allows to observe the dynamics of the superposition of the two intermediate states.

This dynamic can also be understood as interferences between different quantum paths.

Condition to observe interferences:

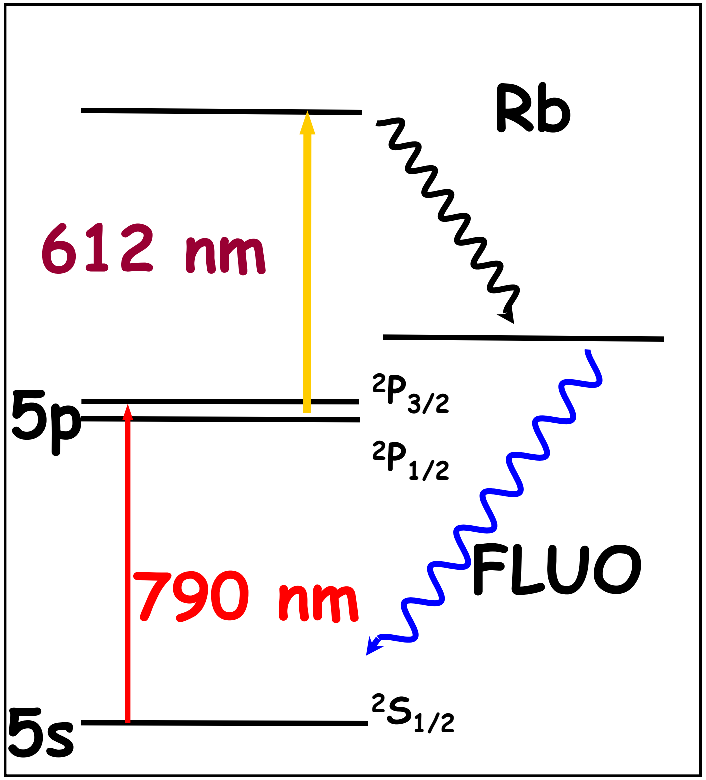
The coupled and uncoupled states should have different probe probabilities

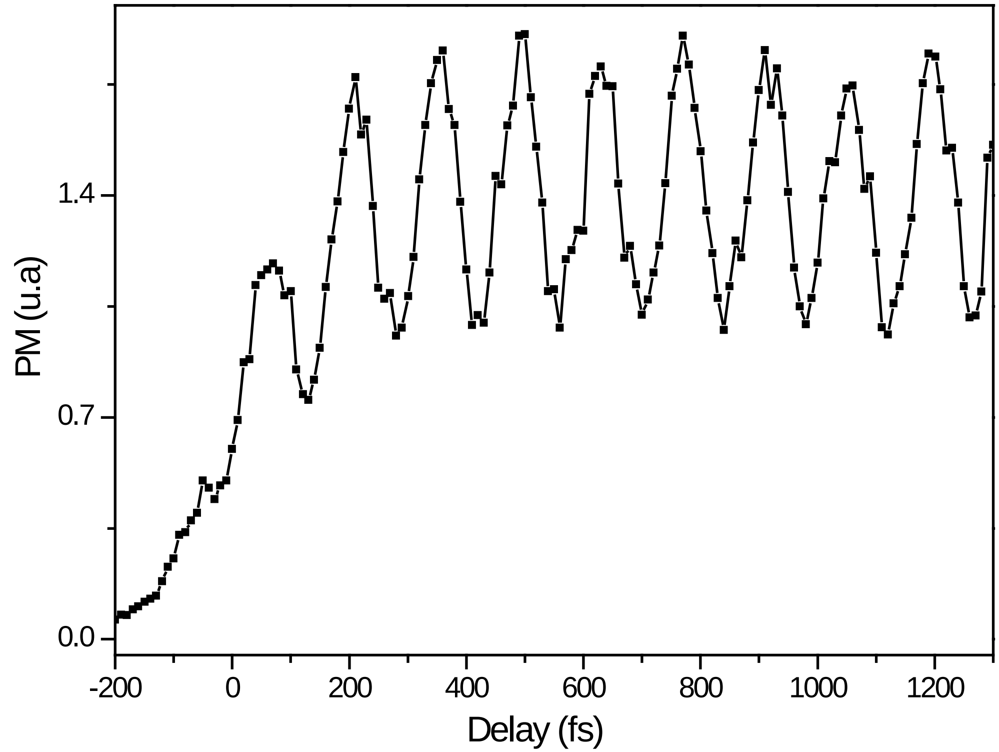




The coupled state evolves freely back and forth towards the uncoupled state.

Spin-orbit precession in Rb



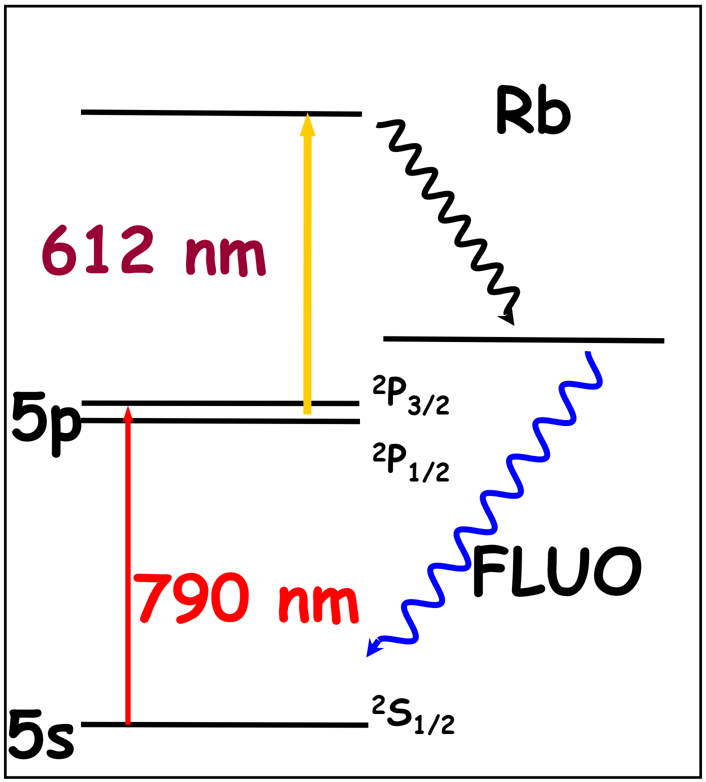


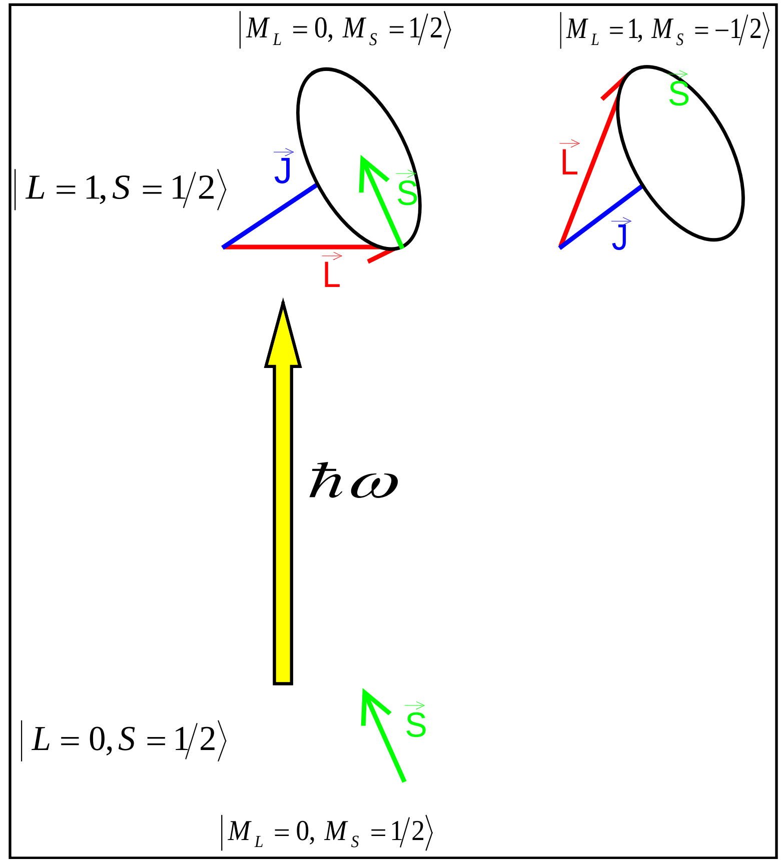
First observed in Potassium then in Rubidium

S. Zamith et al, EPJD 12, 255 (2000)

E. Sokell, et al, JPB 33, 2005 (2000)

Spin-orbit precession in Rb

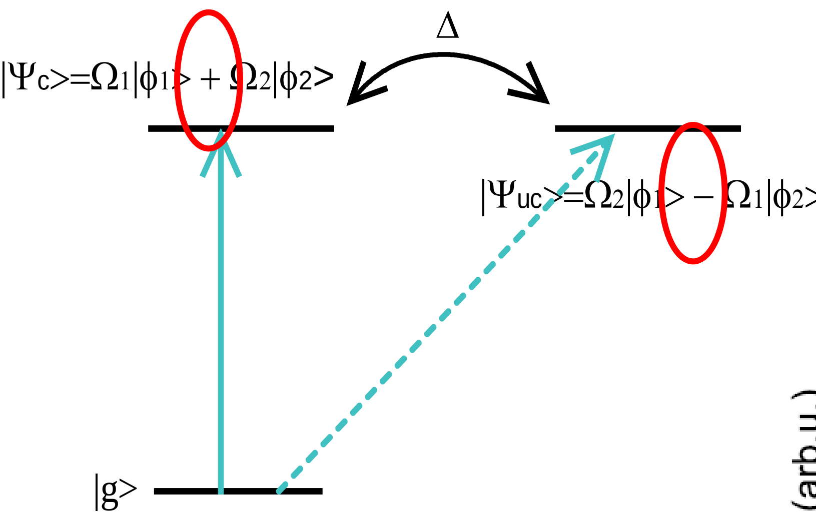




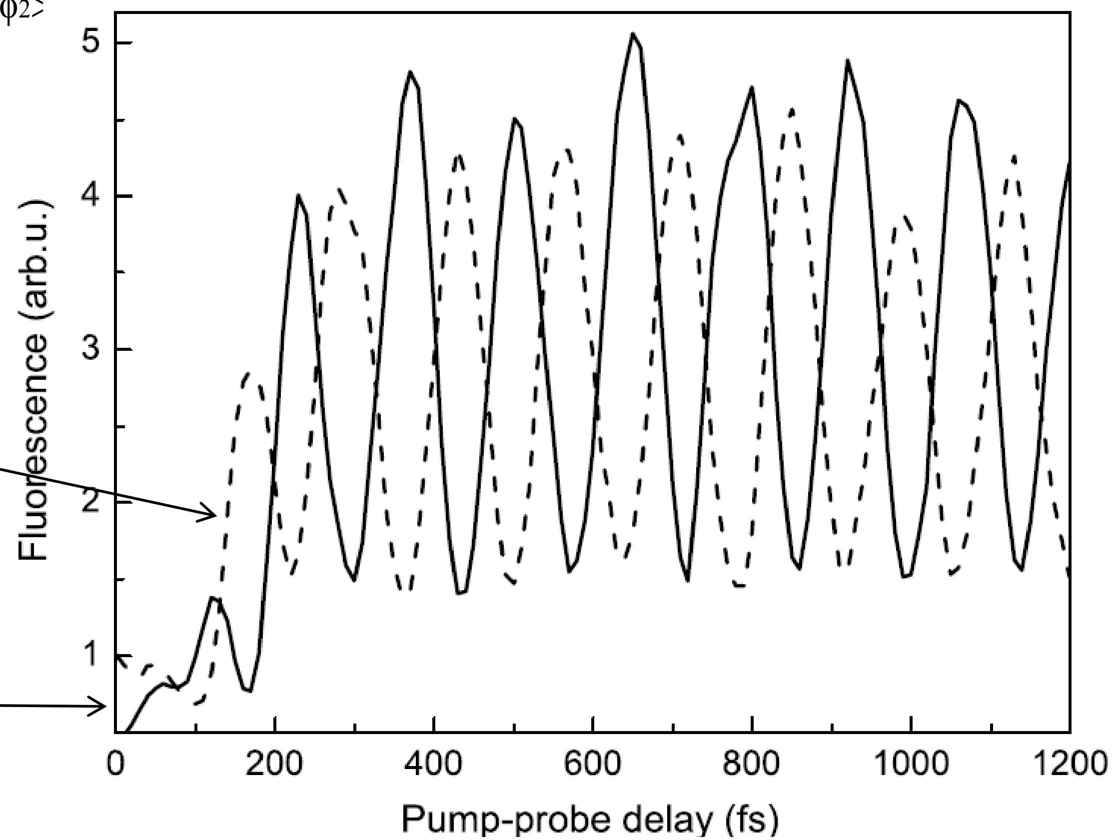
Fine structure : The coupled and uncoupled states belong to the uncoupled basis set.

The spin of the electron is spectator during the pump duration. The free evolution corresponds to the spin precession around the total angular momentum.

Changing the sign of the electric field by applying a π-step



The oscillations of the coupled state are phase shifted by pi with respect to the reference



Without π-step

With π-step

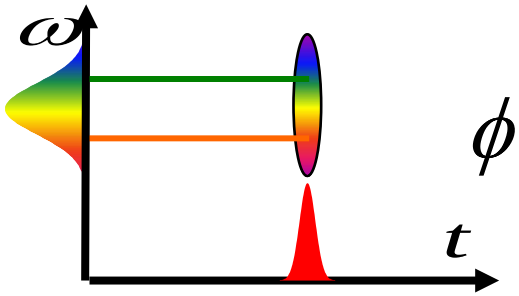
Chatel et al, J Phys B, 41 (2008), 074023

Three examples

- Coherent control to cool molecules
- Coherent control to manipulate spin precession in atoms
- **Coherent transient revisited : Shape the pump and the probe**

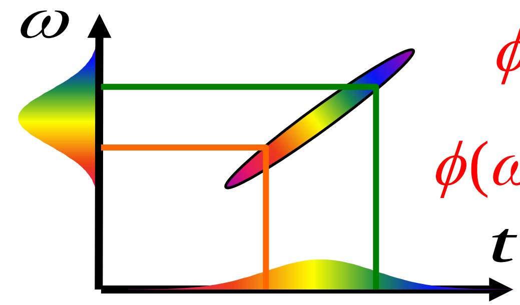
Coherent transients : principle

Fourier limited pulse



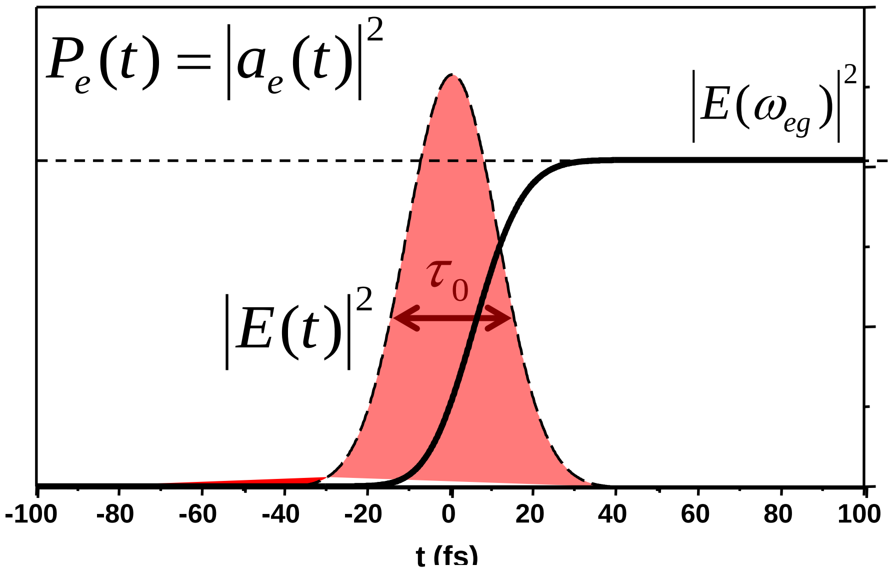
$$\phi(\omega) = 0$$

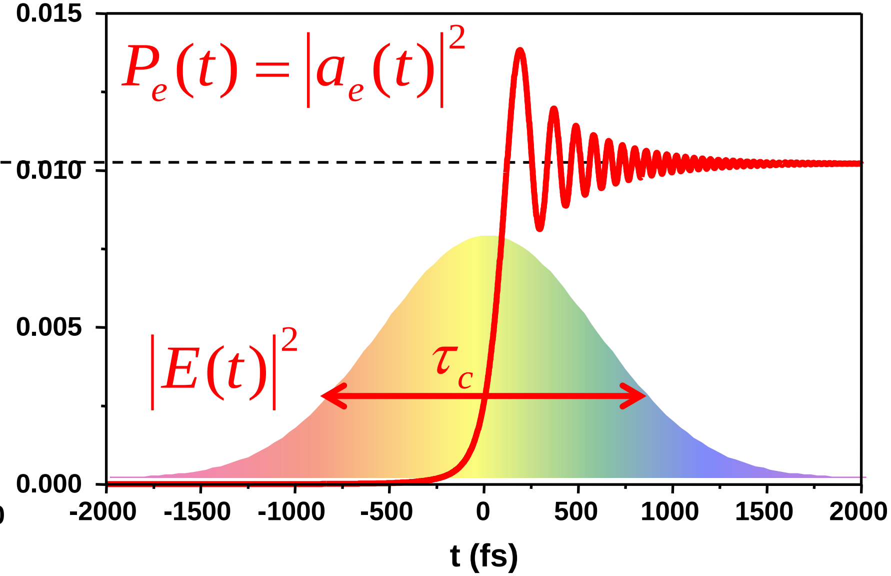
Chirped pulse



$$\phi^{(2)} \gg \tau_0^2$$

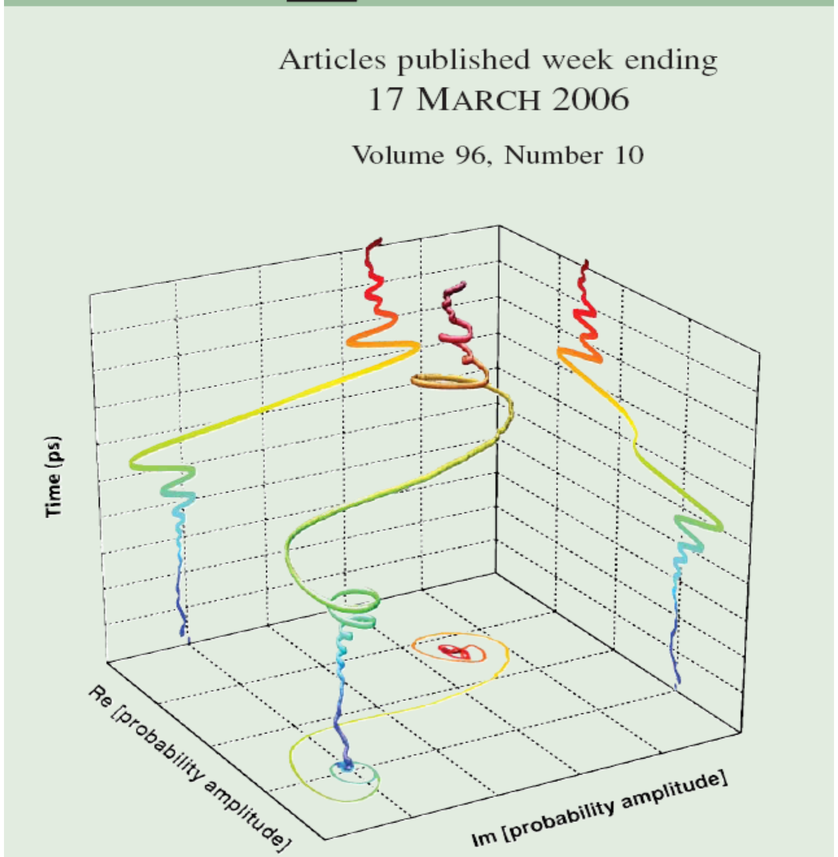
$$\phi(\omega) = \frac{\phi^{(2)}}{2}(\omega - \omega_{eg})^2$$



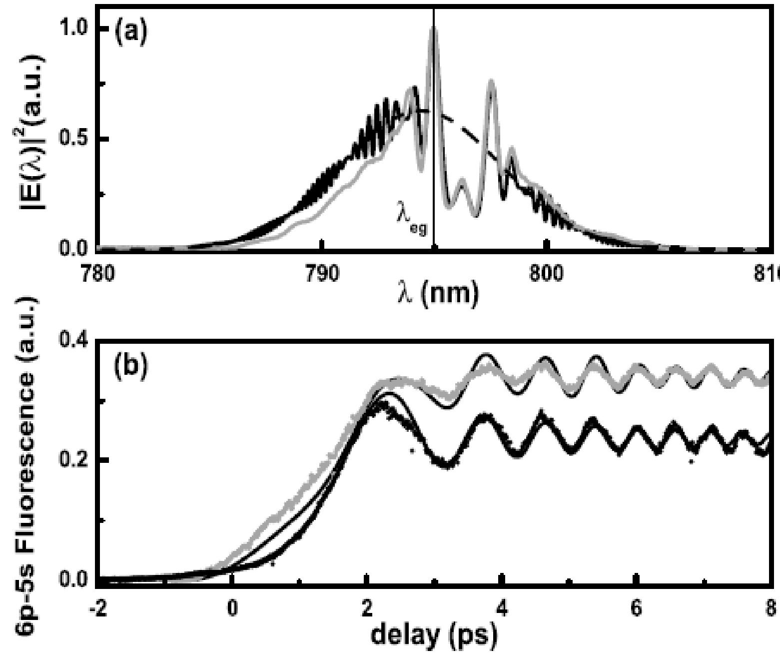


$$a_e(\tau) \propto \int_{-\infty}^{\tau} E(t) e^{i\omega_{eg}t} dt$$

Quantum holography : to follow the wave function in real time



Monmayrant et al
PRL 96, 103002 (2006)



To implement Temporal Fresnel lens

*Degert et al, PRL **89**, 203003-2 (2002)*

To use the coherence of the atom to reconstruct the electric field.

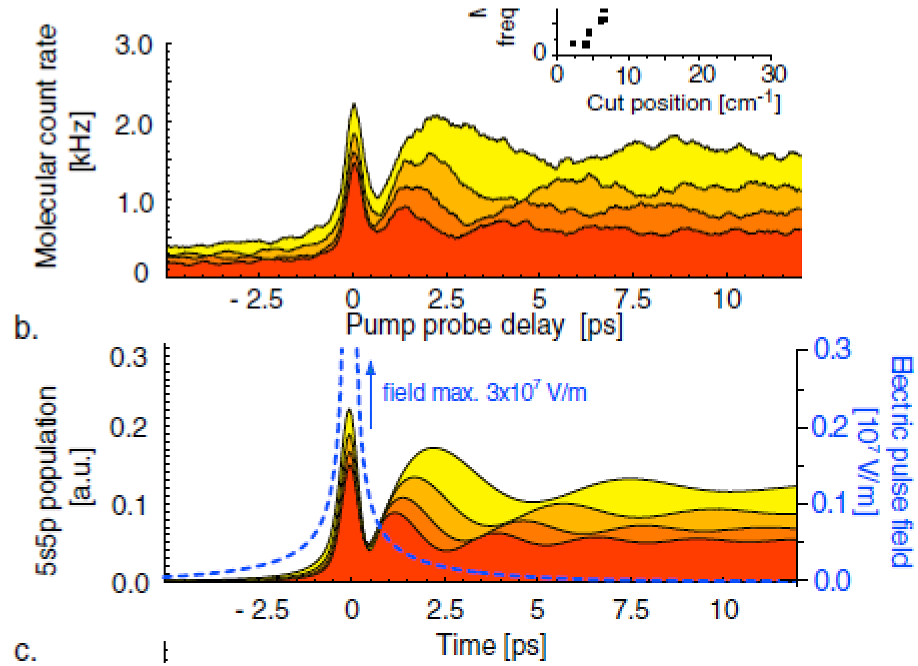
Monmayrant et al OL 31, 410 (2006)

PRL 100, 233003 (2008) PHYSICAL REVIEW LETTERS week ending 13 JUNE 2008

Coherent Transients in the Femtosecond Photoassociation of Ultracold Molecules

W. Salzmann, T. Mullins, J. Eng, M. Albert, R. Wester, and M. Weidemüller*
Physikalisches Institut, Universität Freiburg, Hermann Herder Strasse 3, D-79104 Freiburg im Breisgau, Germany

A. Merli, S. M. Weber, F. Sauer, M. Plewicki, F. Weise, L. Wöste, and A. Lindinger†
Institut für Experimentalphysik, Freie Universität Berlin, Arnimallee 14, D-14195 Berlin, Germany
(Received 11 December 2007; published 13 June 2008)



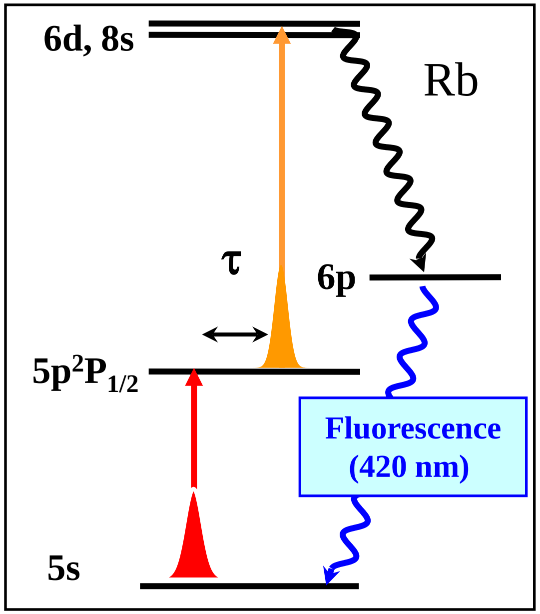
Quantum transients

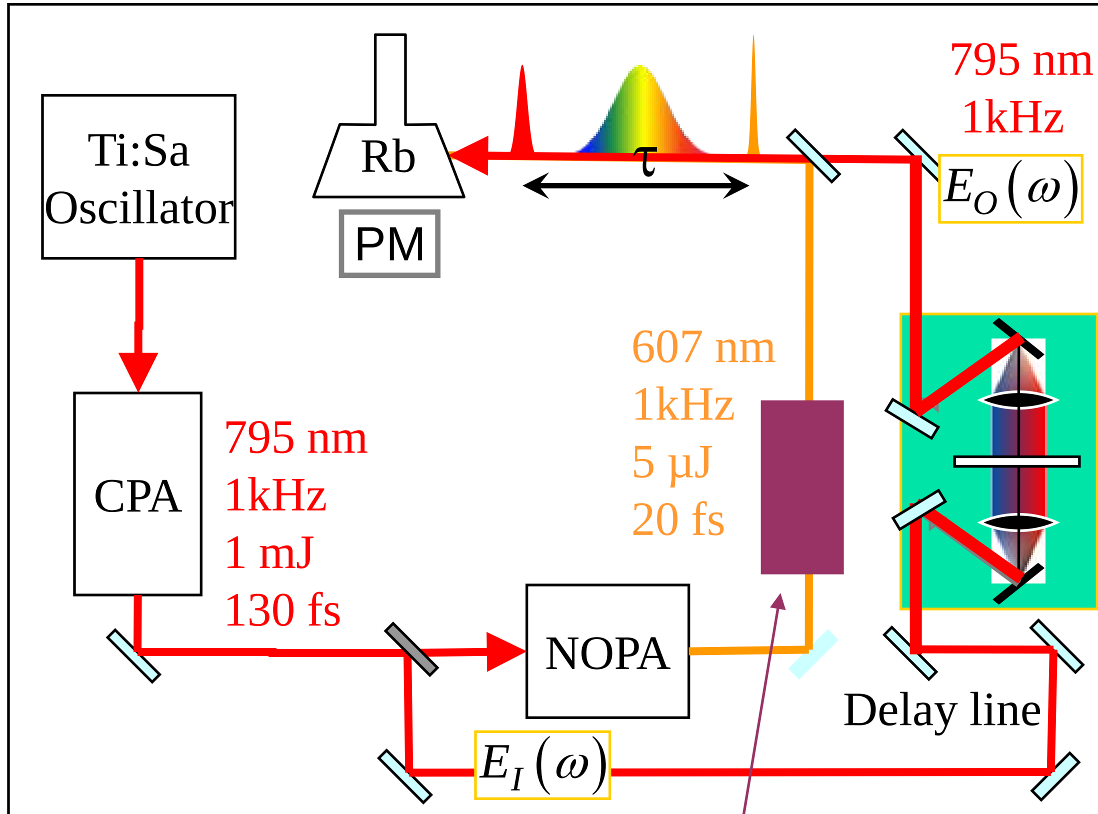
A. del Campo
Institute for Mathematical Sciences, Imperial College London, SW7 2PG, UK
QOLS, The Blackett Laboratory, Imperial College London, Prince Consort Rd., SW7 2BW, UK

G. García-Calderón
Instituto de Física, Universidad Nacional Autónoma de México, Apartado Postal 20 364, 01000 México, D.F., México

J. G. Muga
Departamento de Química-Física, UPV-EHU, Apdo. 644, 48080 Bilbao, Spain

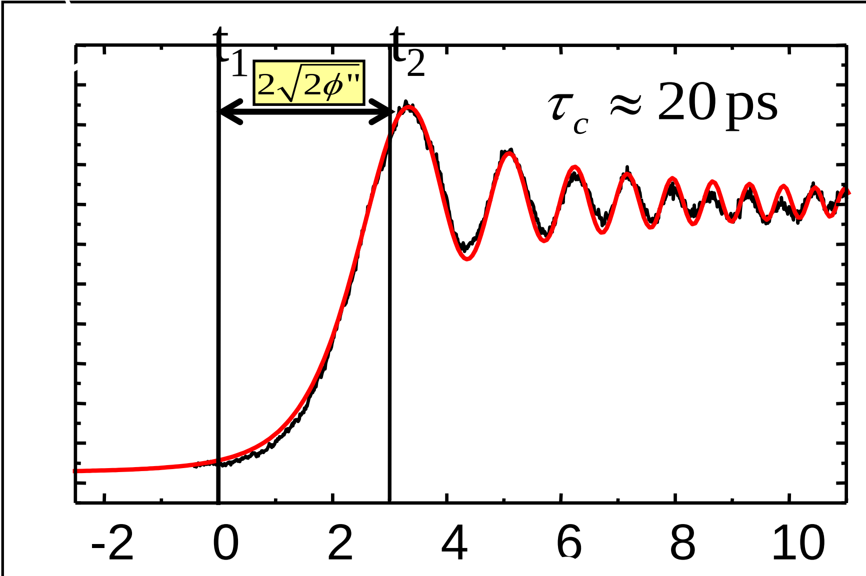
Experimental Set-up



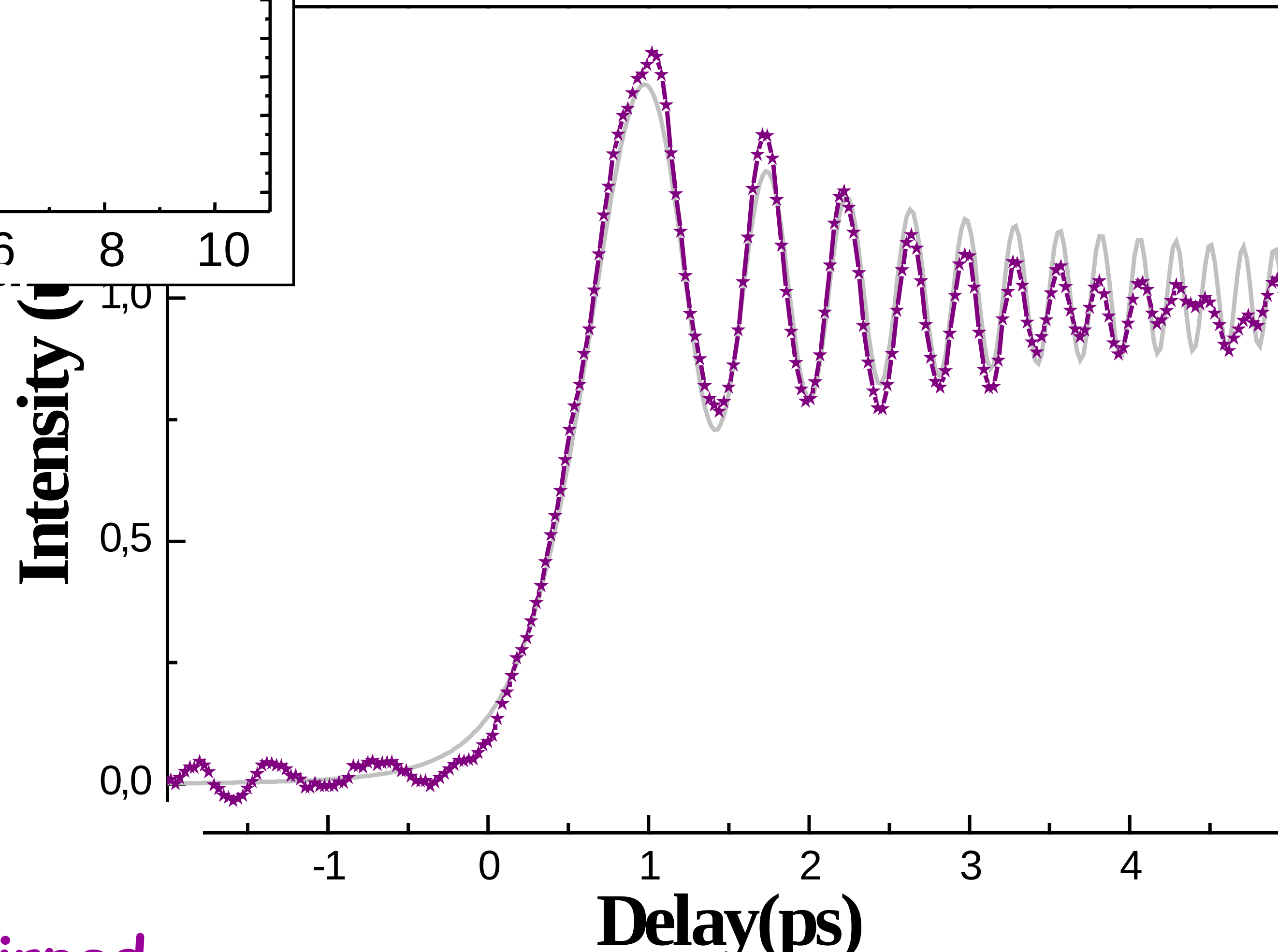


$$E_O(\omega) = H(\omega) E_I(\omega)$$

Grating's pair to chirp the probe.



Pump strongly chirped/probe TF limited

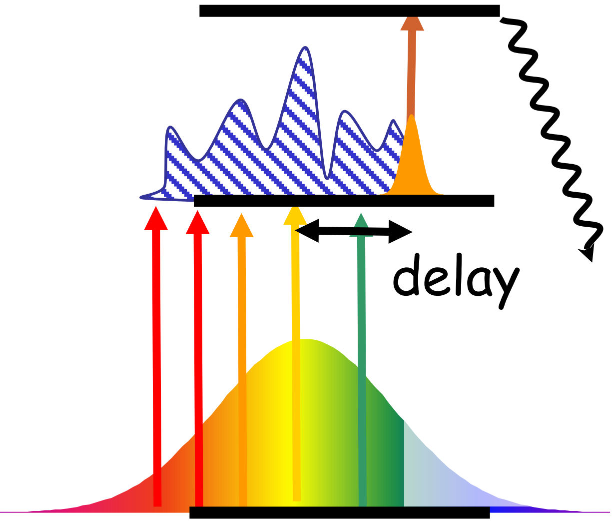


Pump TF limited

Probe strongly chirped

chirp=-1.4 10^5 fs^2

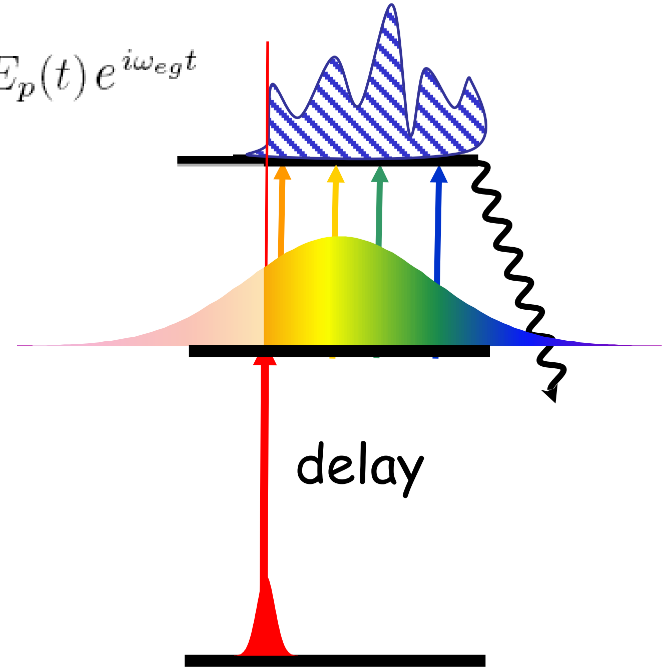
$$b_f(\tau) = -\frac{\mu_{fe}\mu_{eg}}{4\hbar^2}\int_{-\infty}^{\infty} dt' E_s(t')\, e^{i\omega_{fe}t'} \int_{-\infty}^{t'+\tau} dt\, E_p(t)\, e^{i\omega_{eg}t}$$



Chirped pump

Short probe

$$b_f(\tau) \propto \int_{-\infty}^{\tau} E_{pump}(t) e^{i\omega_{eg}t} dt$$



Short pump

Chirped probe

$$b_f(\tau) \propto \int_{\tau}^{+\infty} E_{probe}(t) e^{i\omega_{fe}t} dt$$

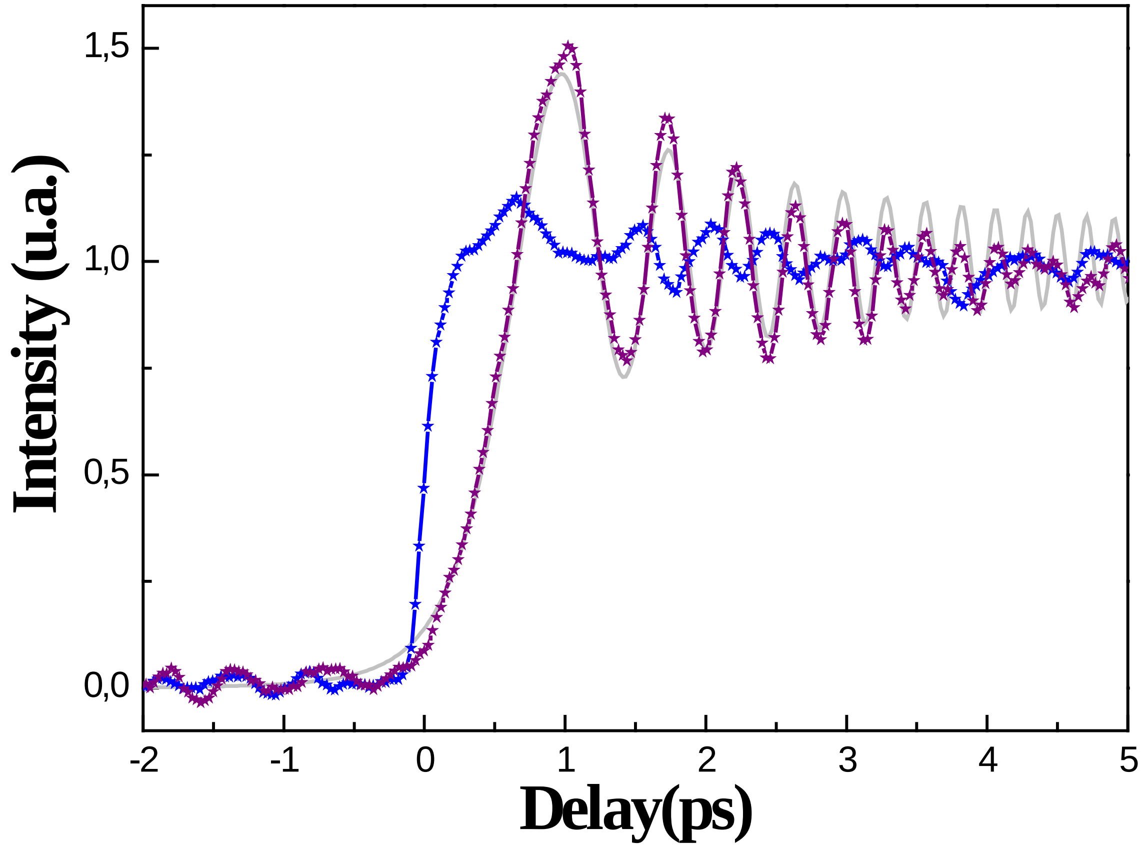
- If now we apply an opposite chirp on the pump we are able to suppress the coherent transients

Pump strongly chirped

Phisec=1.4 10^5 fs^2

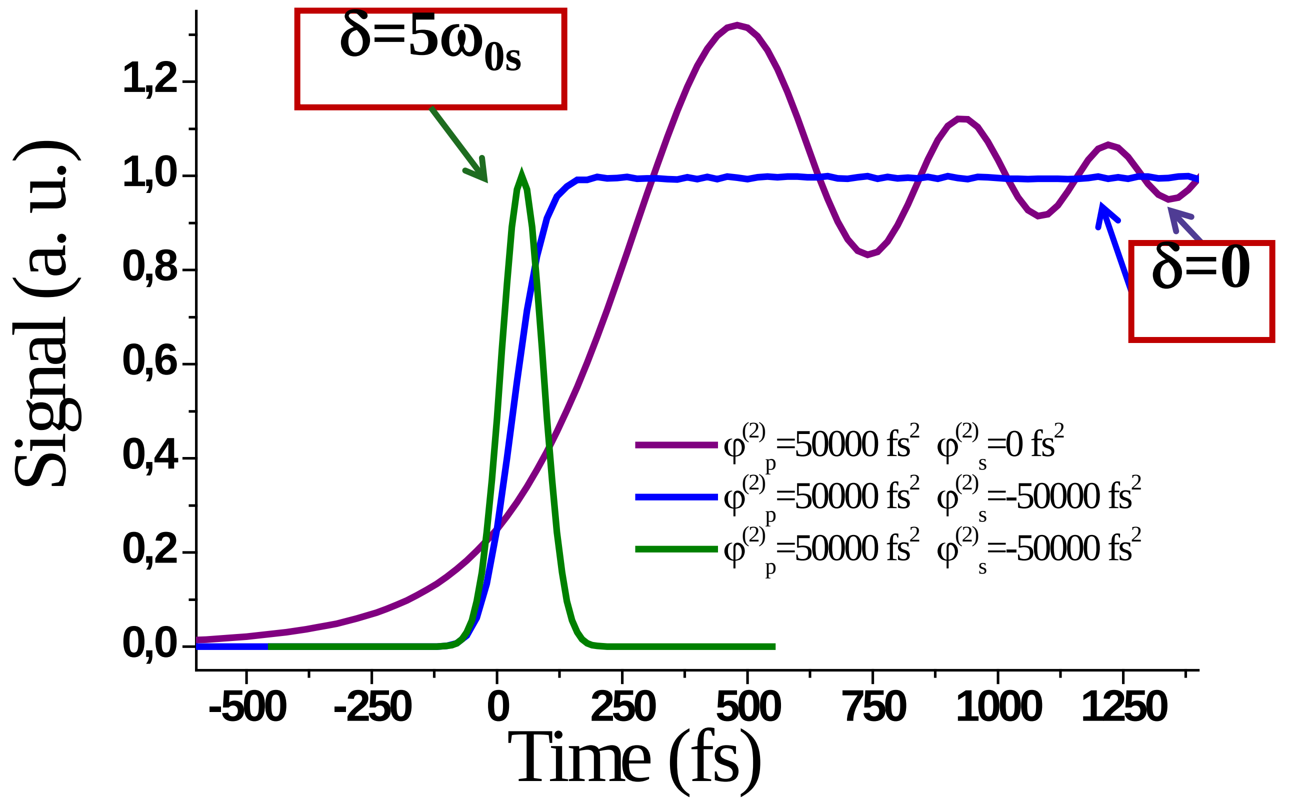
Probe strongly chirped

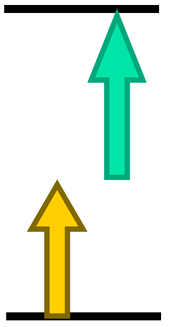
Phisec=-1.4 10^5 fs^2



Another way to characterize pulse in an unusual spectral range

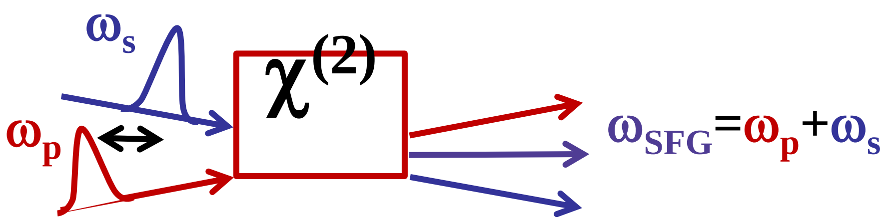
Short dynamic even pulses are long



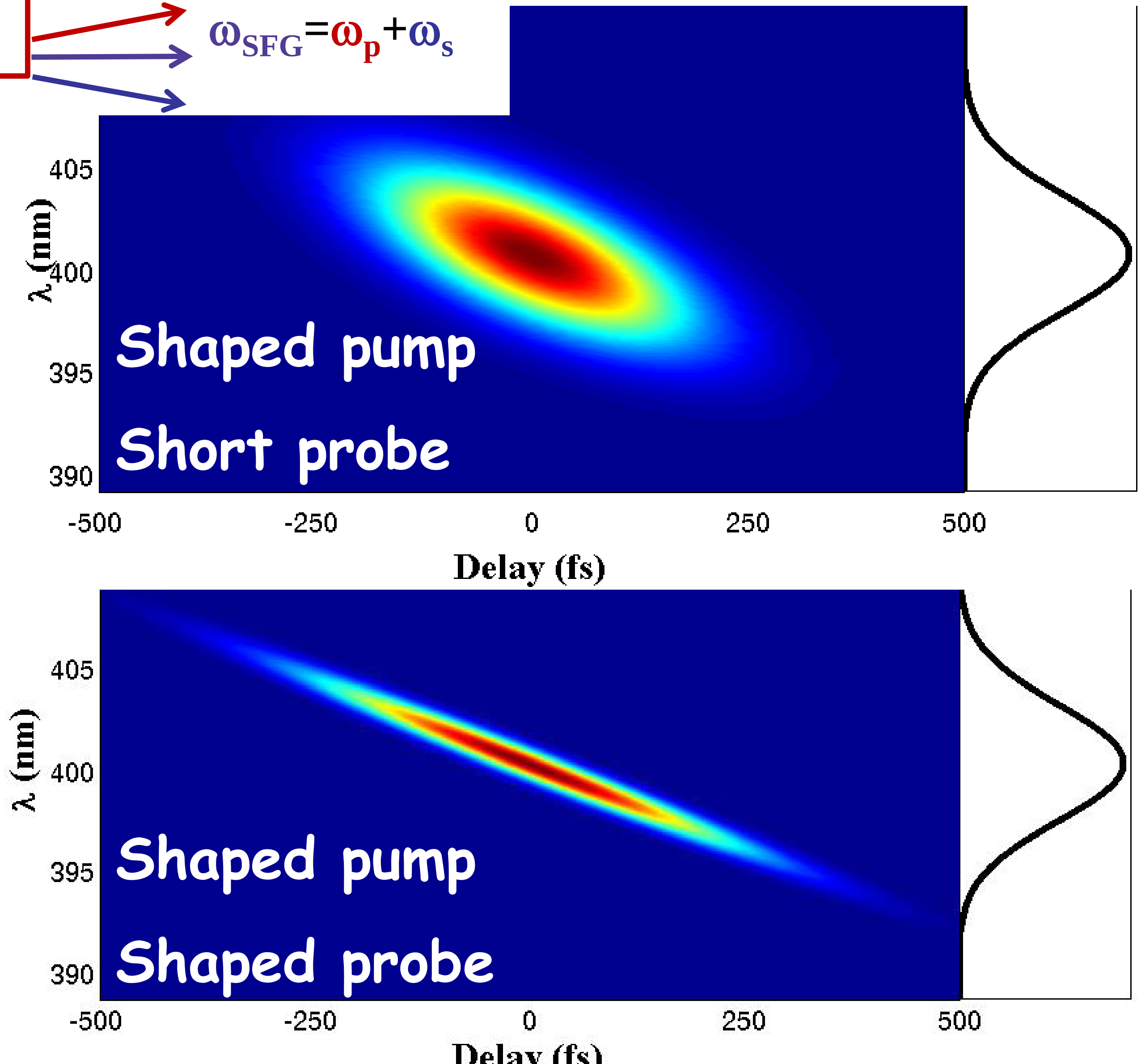


Case at resonance or without any intermediate state.

Preliminary work Using sum frequency mixing

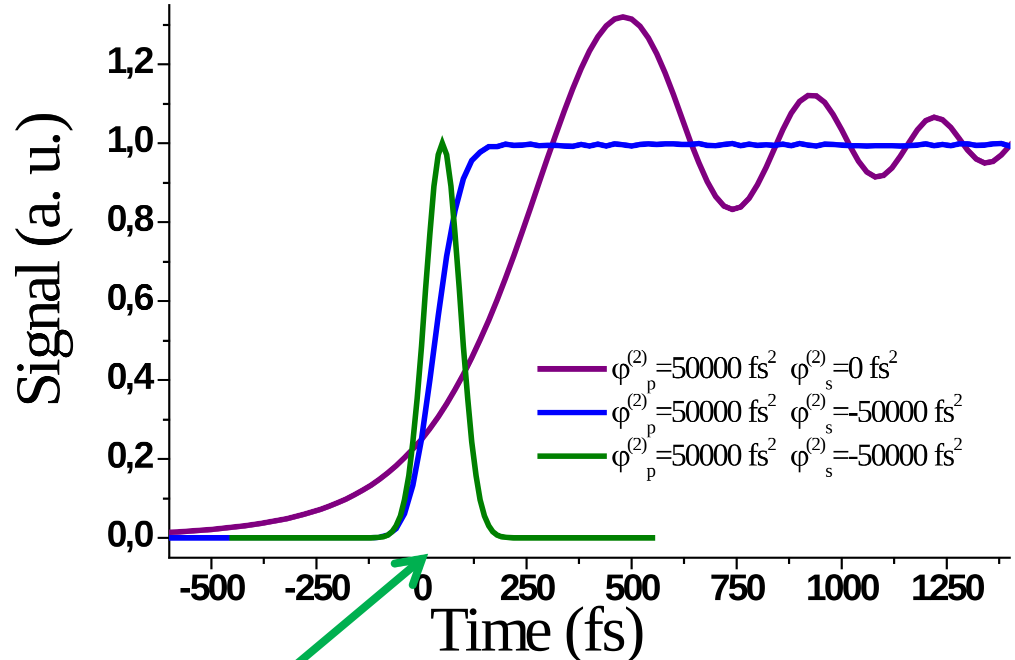


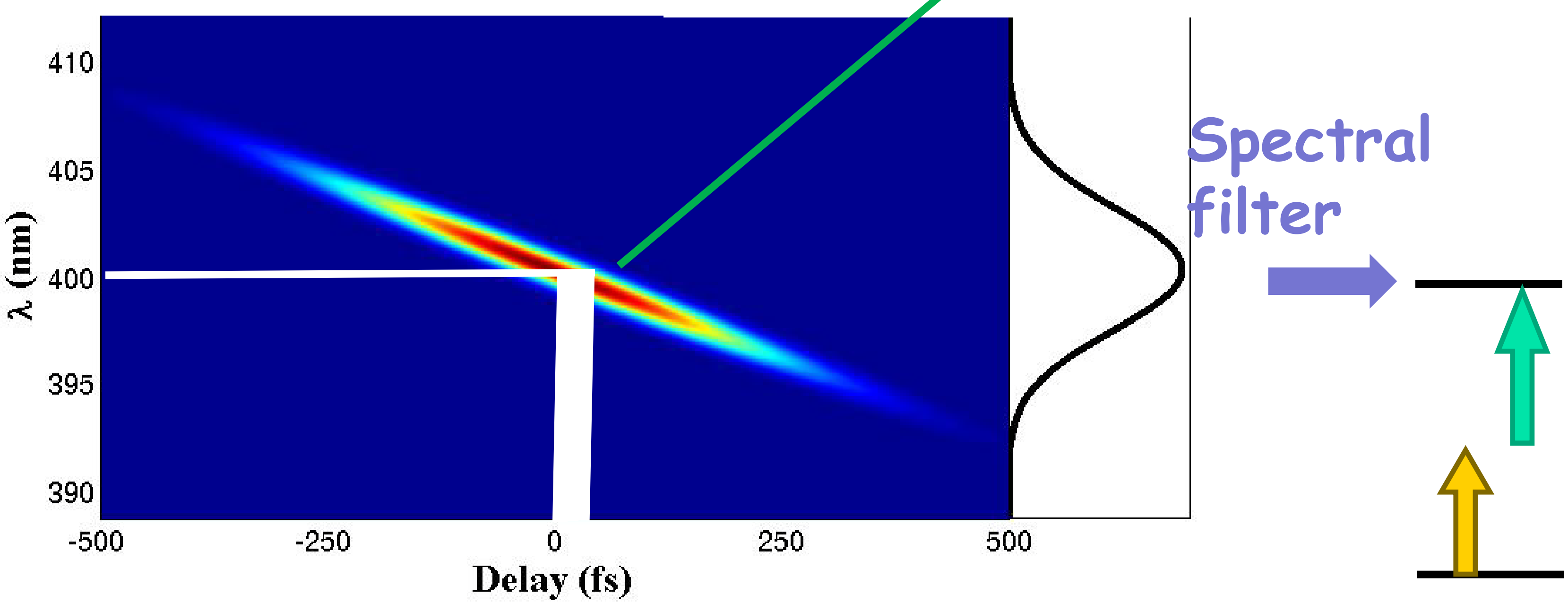
Case of a quadratic phase

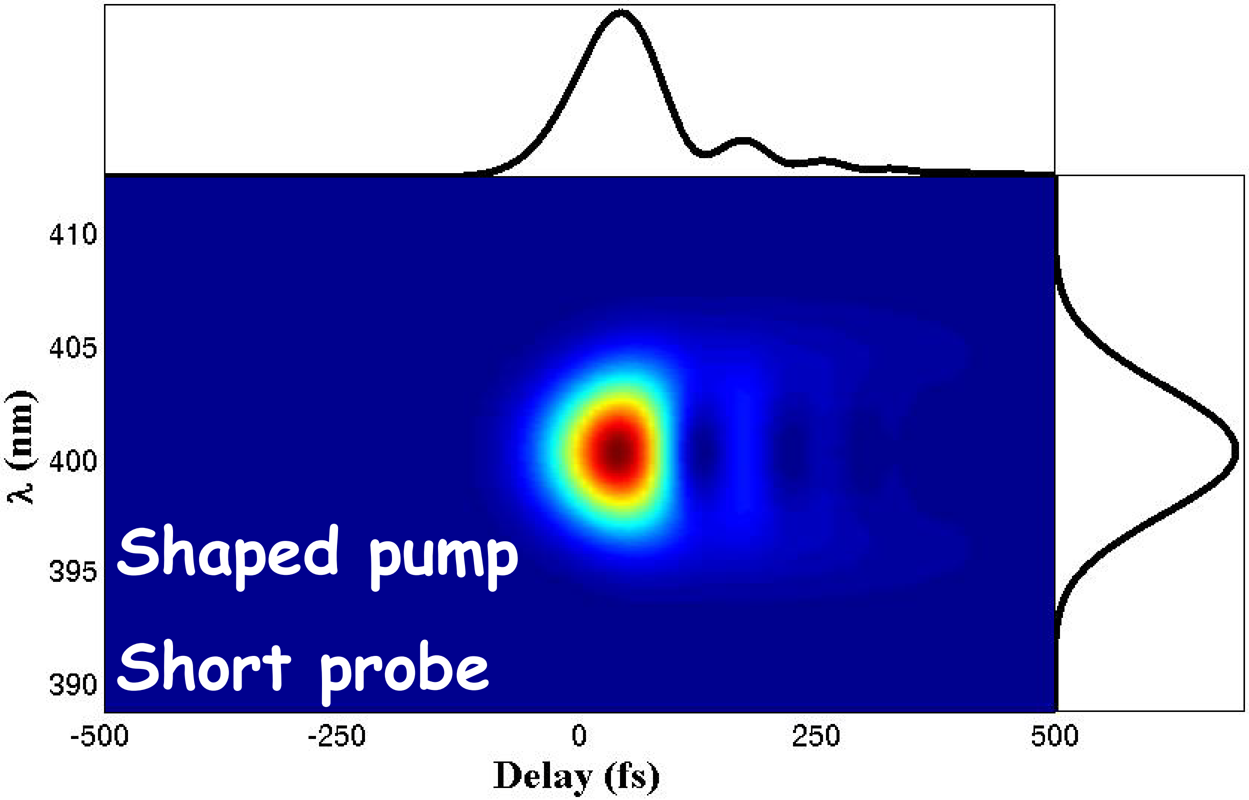


Femto grou

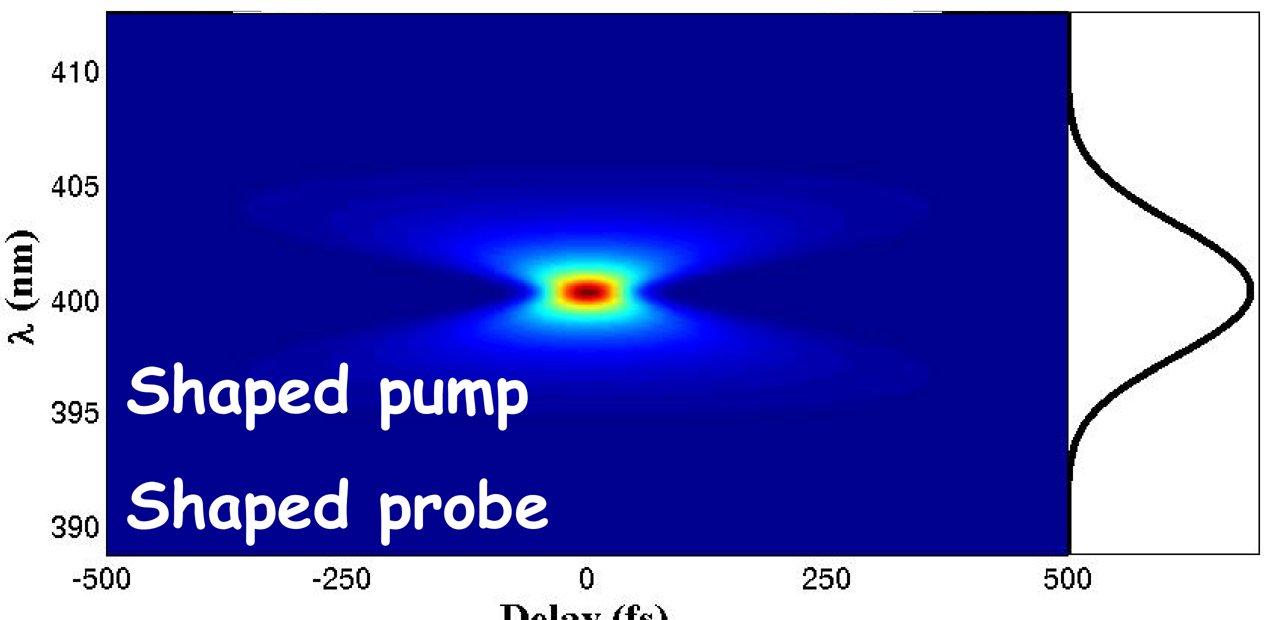
The final state acts as a spectral filter







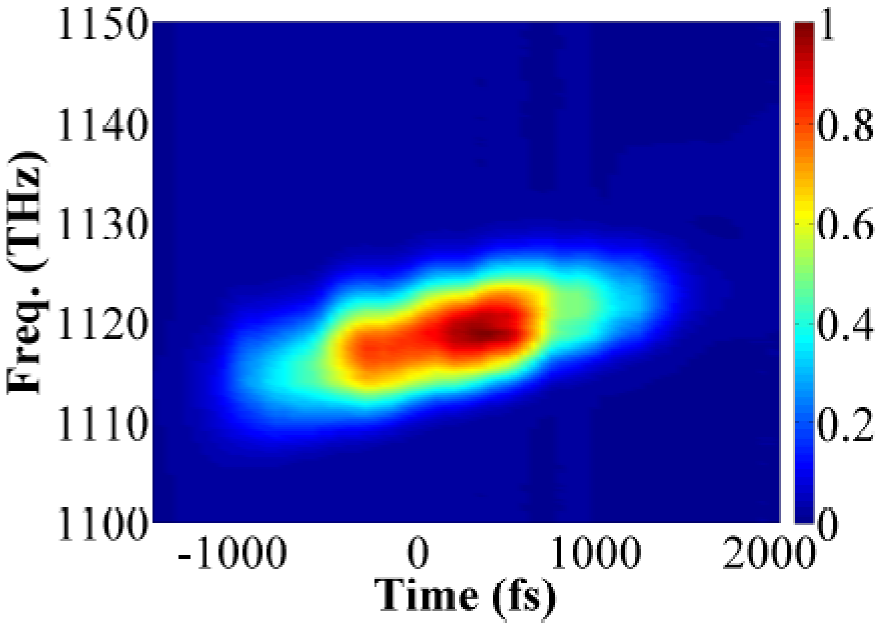
Case of a cubic phase

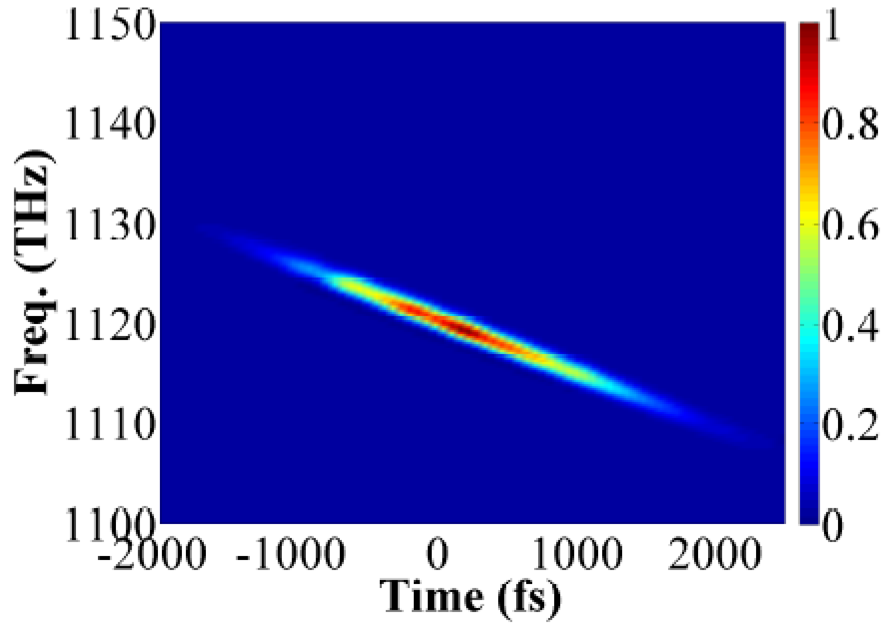


A sort of focus point

Both in time and spectral domain!

Work in progress





- Experiment in progress
- A new spectroscopic approach
- Investigation to use HHG as either the pump or the probe (in coll P. Salières -CEA)

Conclusion

- Coherent control : a way to cool molecules
- Coherent control : To manipulate the spin by using shaped pulses
- To shape pump and probe could lead to a new spectroscopic approach : work under study
- FASTQUAST European network

(PhD and postdoc positions)

http://icb.u-bourgogne.fr/fastquast/