High resolution cavity-enhanced absorption spectroscopy with a mode comb.

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Research at LSP/Grenoble

Spectroscopy

• molecules of amospheric interest (O_3, CH_4, CO_2…) : ICLAS 0.6-1.1µm, CW-CRDS in the telecom range — Alain Campargue, Samir Kassi (poster)

• Isotope effects (in NO_2) : LIF, CRDS… on a supersonic jet — Remy Jost

Trace detection

• Near-mid IR : DFB diode lasers & OF-CEAS — Marc Chenevier

• Blue spectral range ECDL diode + OF-CEAS — Irene Courtillot (poster)

New developments…

Mode-Comb CEAS : Detecting radicals in the UV ? — ...

VECSEL lasers (~2.5µm) — Daniele Romanini

Introduction

• Several CRDS/CEAS schemes exist based on different light sources.

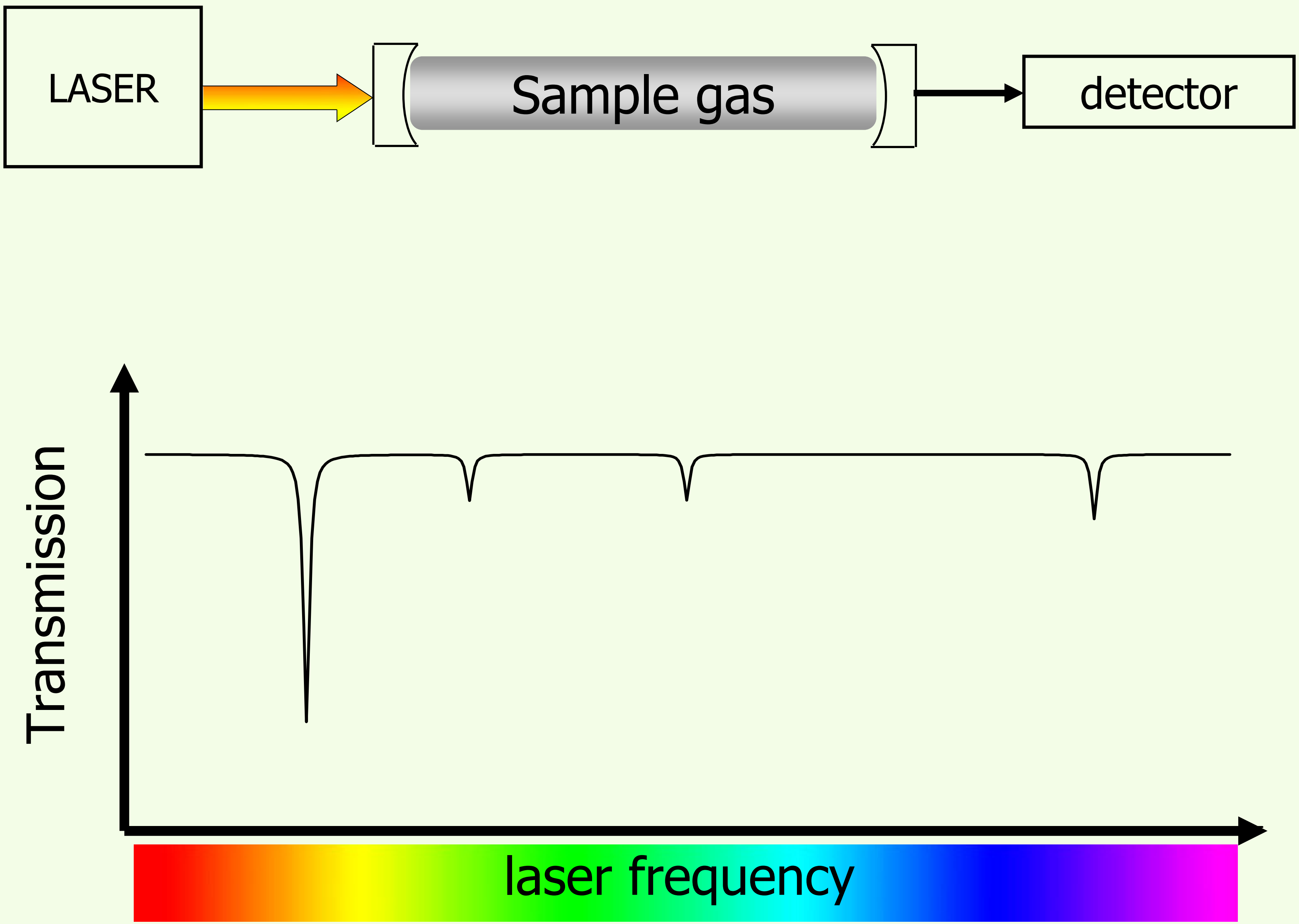
• Increasing interest for Broad Band coverage for several applications. Pulsed dye lasers, lamps, LEDs, and mode-locked femtosecond sources being tested.

• BB CEAS/CRDS: Need to do a compromise dispersion/luminosity: The more you disperse, the less light you have per spectral element, the longer you have to accumulate.

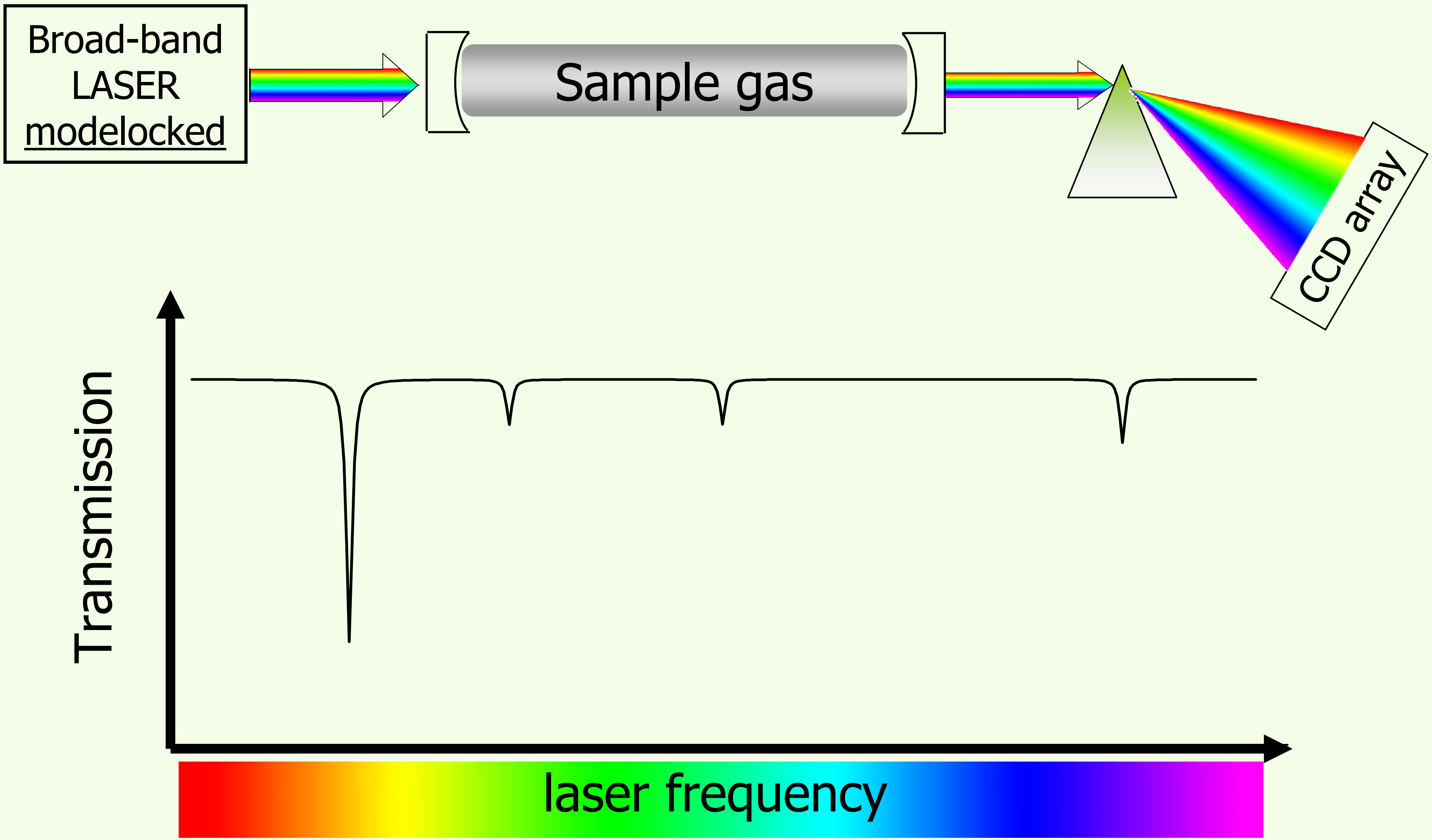
• Other problem is spectral quality and stability of the BB source: Dye lasers and lamps are not very good, contrary to LEDs and ML lasers.

• ML lasers have high beam quality (mode matching…), and adjustable spectrum (width & spectral range) by using nonlinear optics (fibers, etc)

Standard CEAS scheme with laser tuning

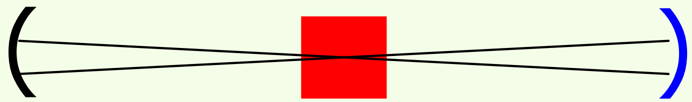


Wavelength Multiplexed Spectroscopy

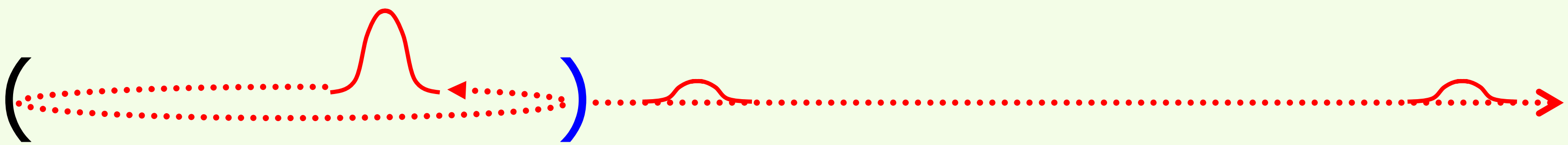


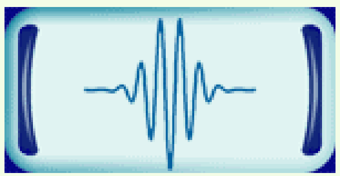
What is a mode-locked laser ?

Laser cavity (very simplified!)



Laser at work…

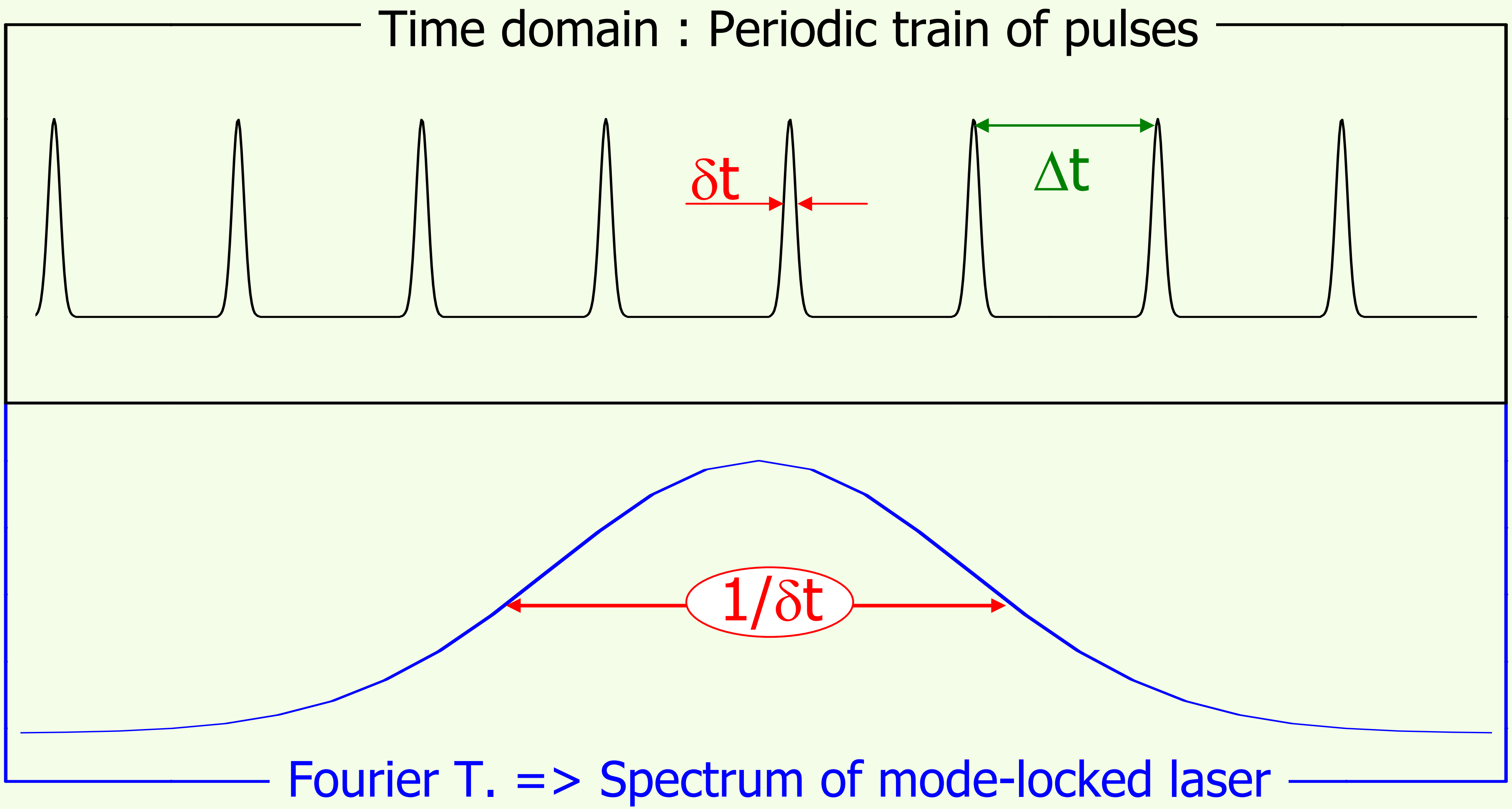




http://www.mpq.mpg.de/~haensch/comb/research/combs.html

Spectrum of a mode-locked laser

... coherent ensemble of single-frequency lasers



in a real case (laser Ti:Sa) the spectrum has $\sim 10^5$ modes

Spectrum of a mode-locked laser

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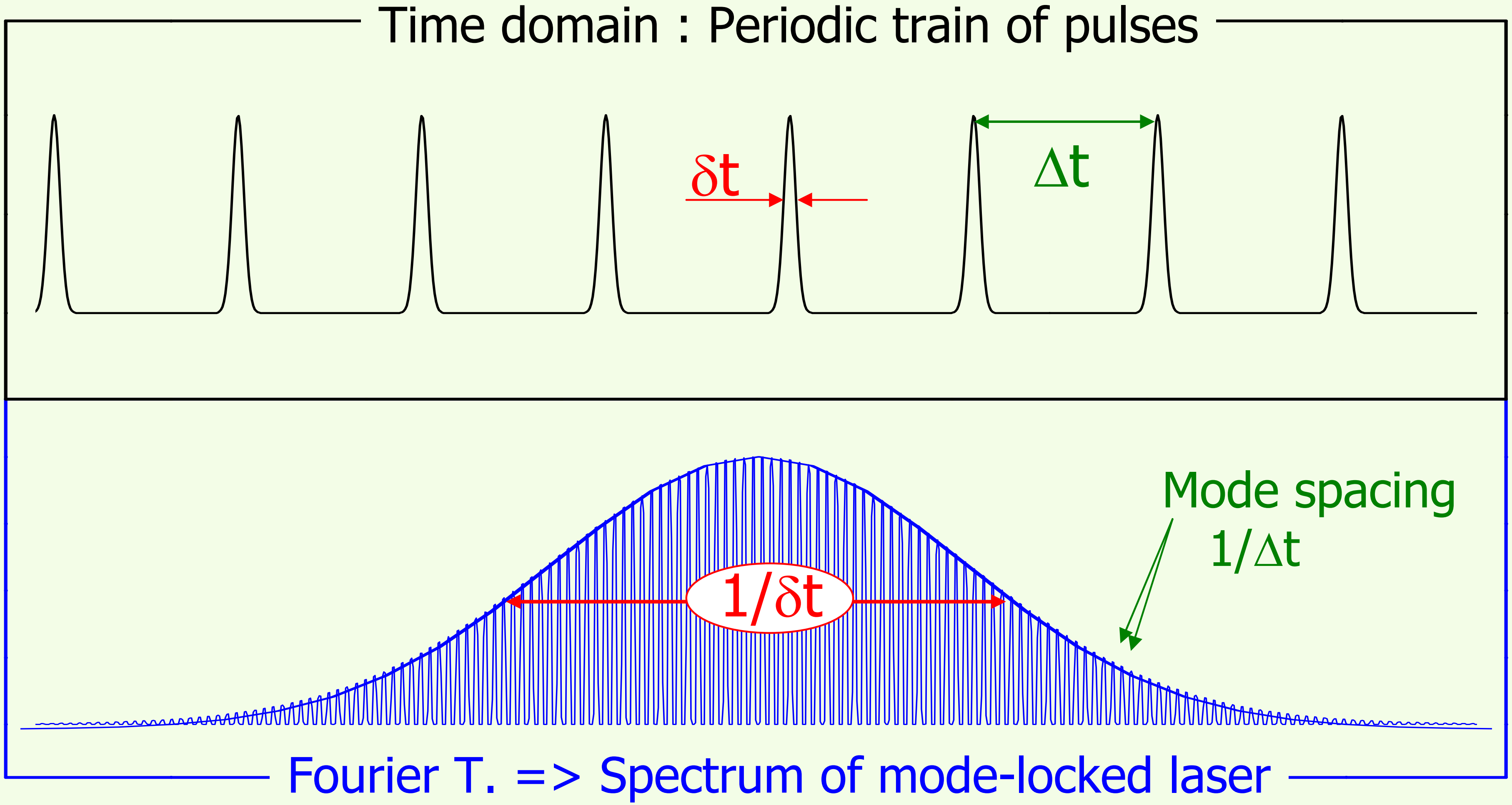
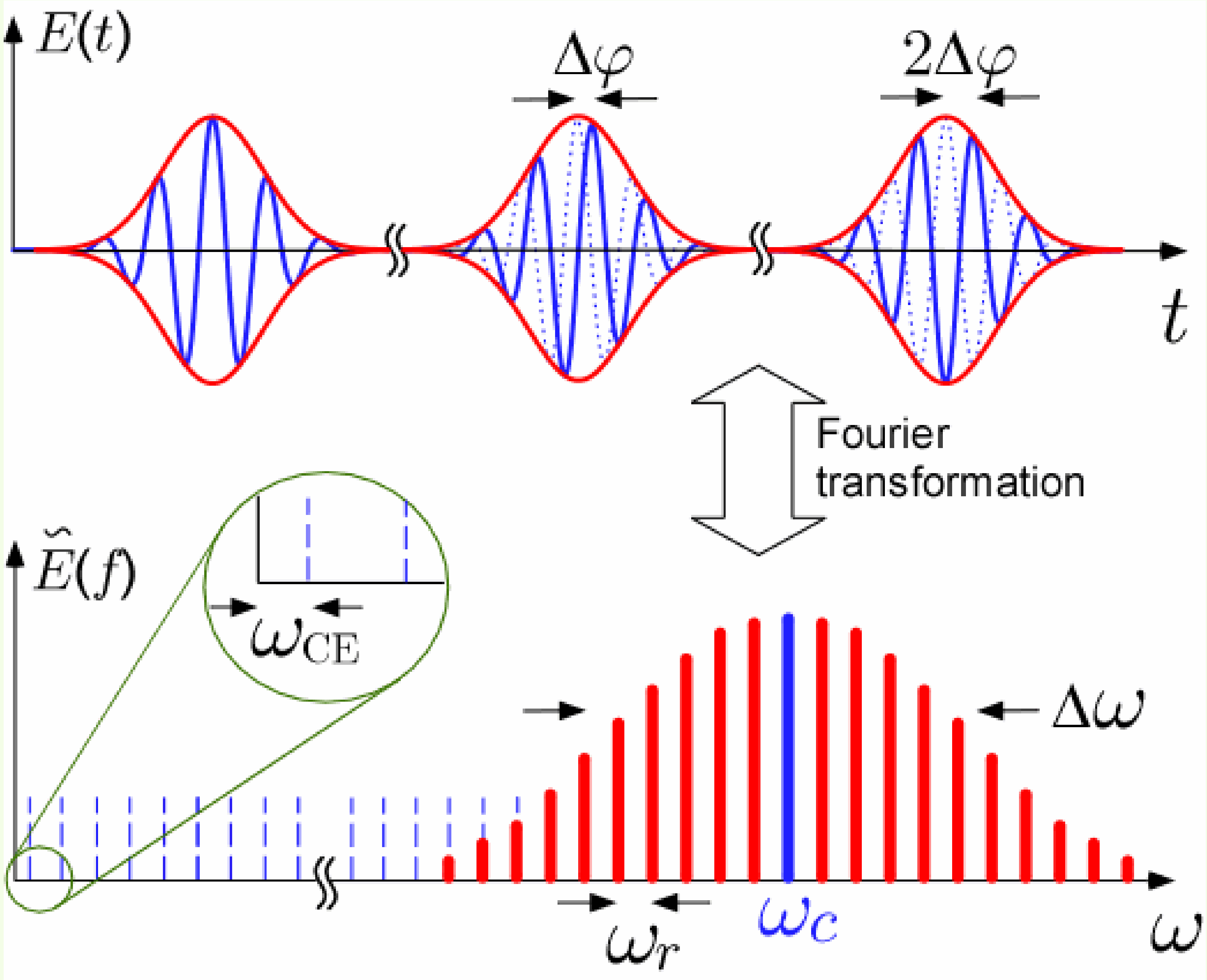


in a real case (laser Ti:Sa) the spectrum has ~10^5 modes

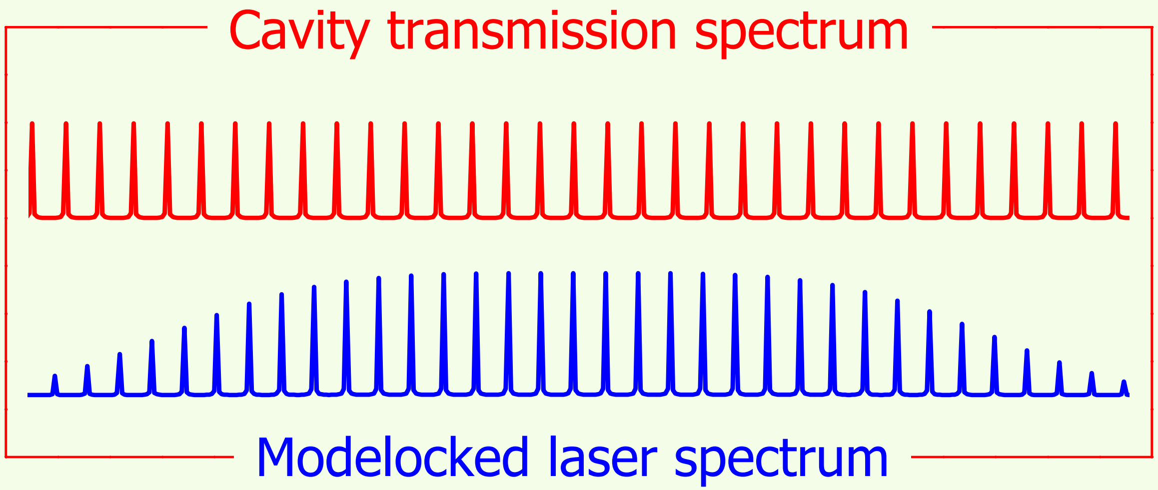
Spectrum of a mode-locked laser

- A closer look -

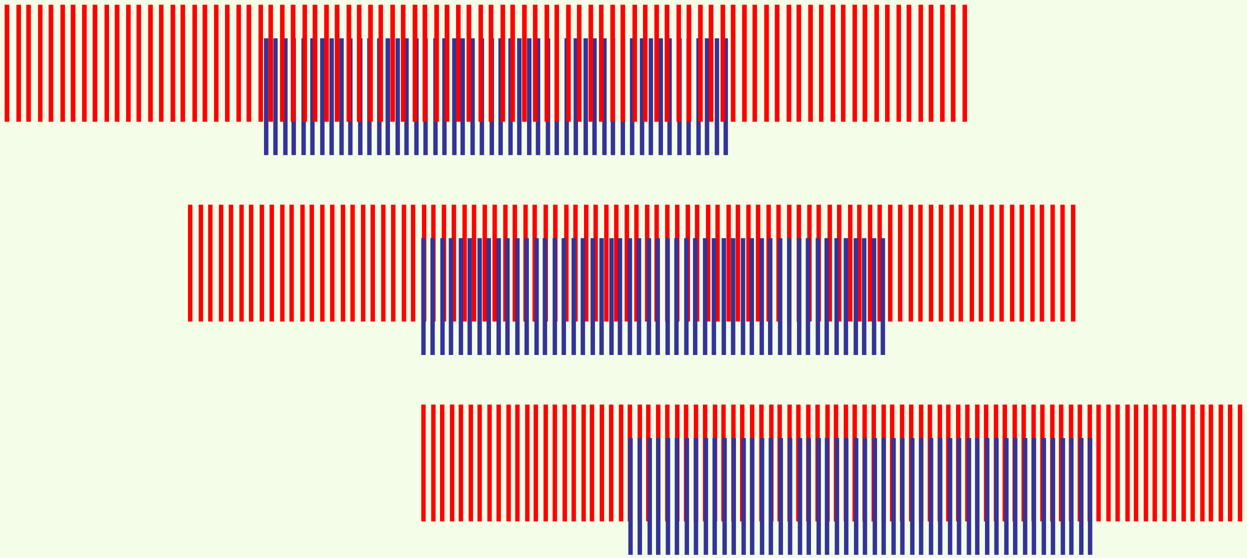


http://www.mpq.mpg.de/~haensch/comb/research/combs.html

"ML-CEAS" : Basic principle



Output Spectrum = T_{cav} x Input Spectrum



The spectrum transmitted by the cavity depends on the overlap of the two combs of modes :

by changing the cavity length, we observe comb beatings, with different periods…

When combs are in tune, the laser spectrum is efficiently transmitted without beatings…

=> " Magic point "

Pulse stacking : The time-domain point of view

Pulses overlap in phase with circulating intra-cavity pulse :

buildup occurs and transmission is efficient !

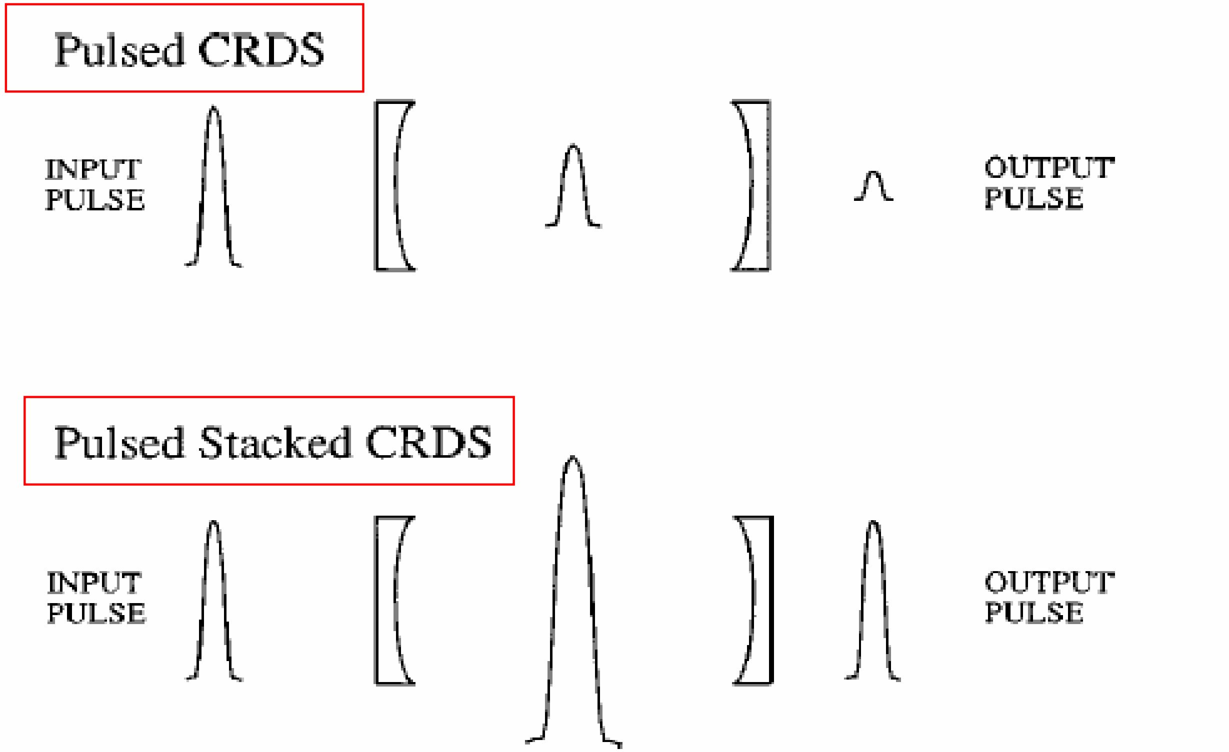
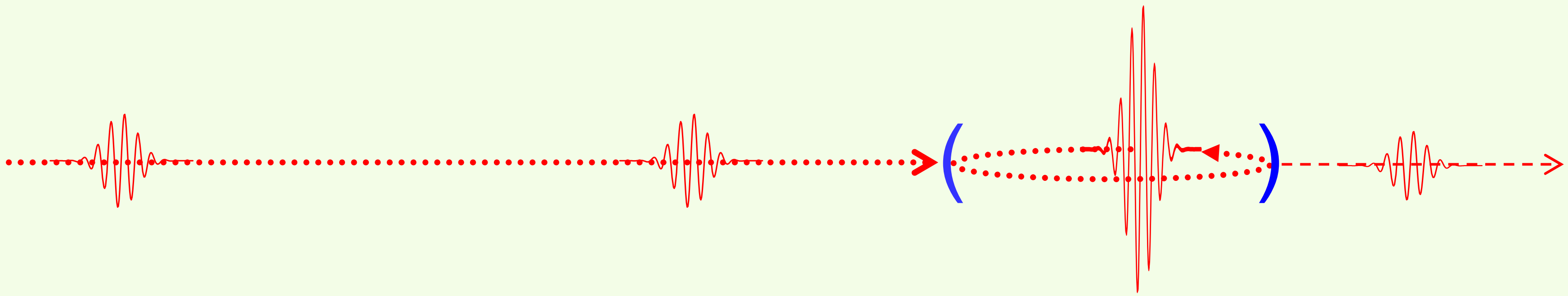


FIG. 1. Illustration of the difference between pulsed CRDS and pulse-stacked CRDS. For cavity mirrors with 500 ppm transmission, the transmitted energy for pulsed CRDS is $(2.5\times10^{-7})U_{\text{inc}}$. For pulse-stacked CRDS the pulse energy inside the RDC is $(2\times10^{3})U_{\text{inc}}$ and the transmitted energy is U_{inc}.

E. R. Crosson et al., REV. SCI. INSTRUM. 70 (1999) 4

An alternative point of view in the time domain…

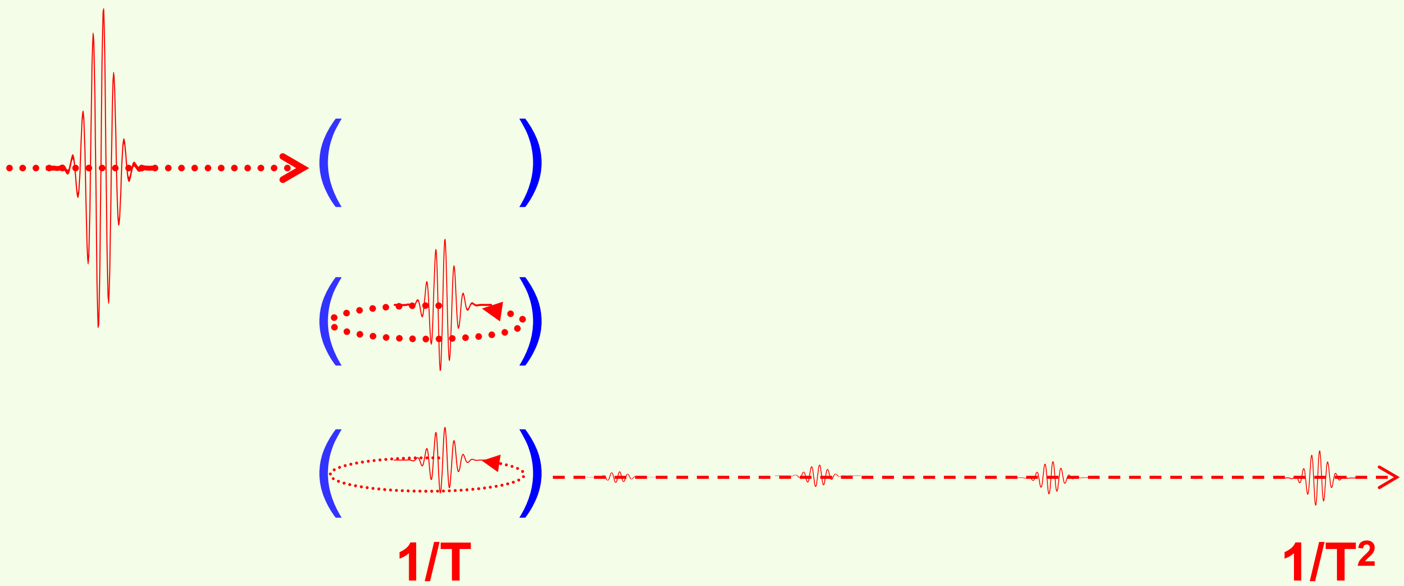
Laser pulses fall in phase with the pulse circulating in the cavity, which gives a coherent buildup of the intracavity field. This is similar to the single-frequency case (CW-CRDS) but the buildup is not a stationary wave…



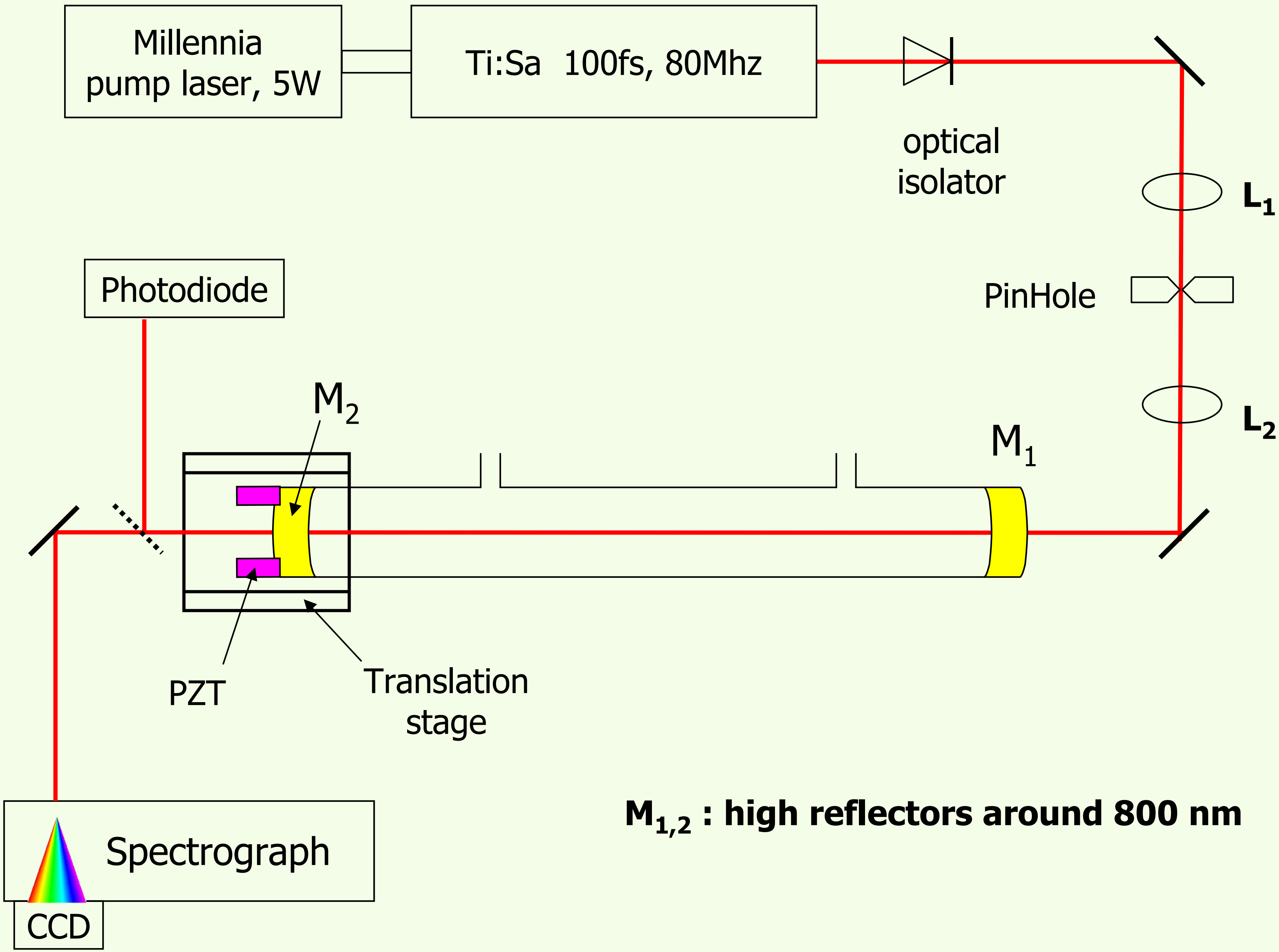
Comparison with the case of a lonely pulse

No coherent buildup, instead a decreasing sequence of output pulses is produced (ring-down...), with initial intensity < T^2

Cavity injection is much less effective...

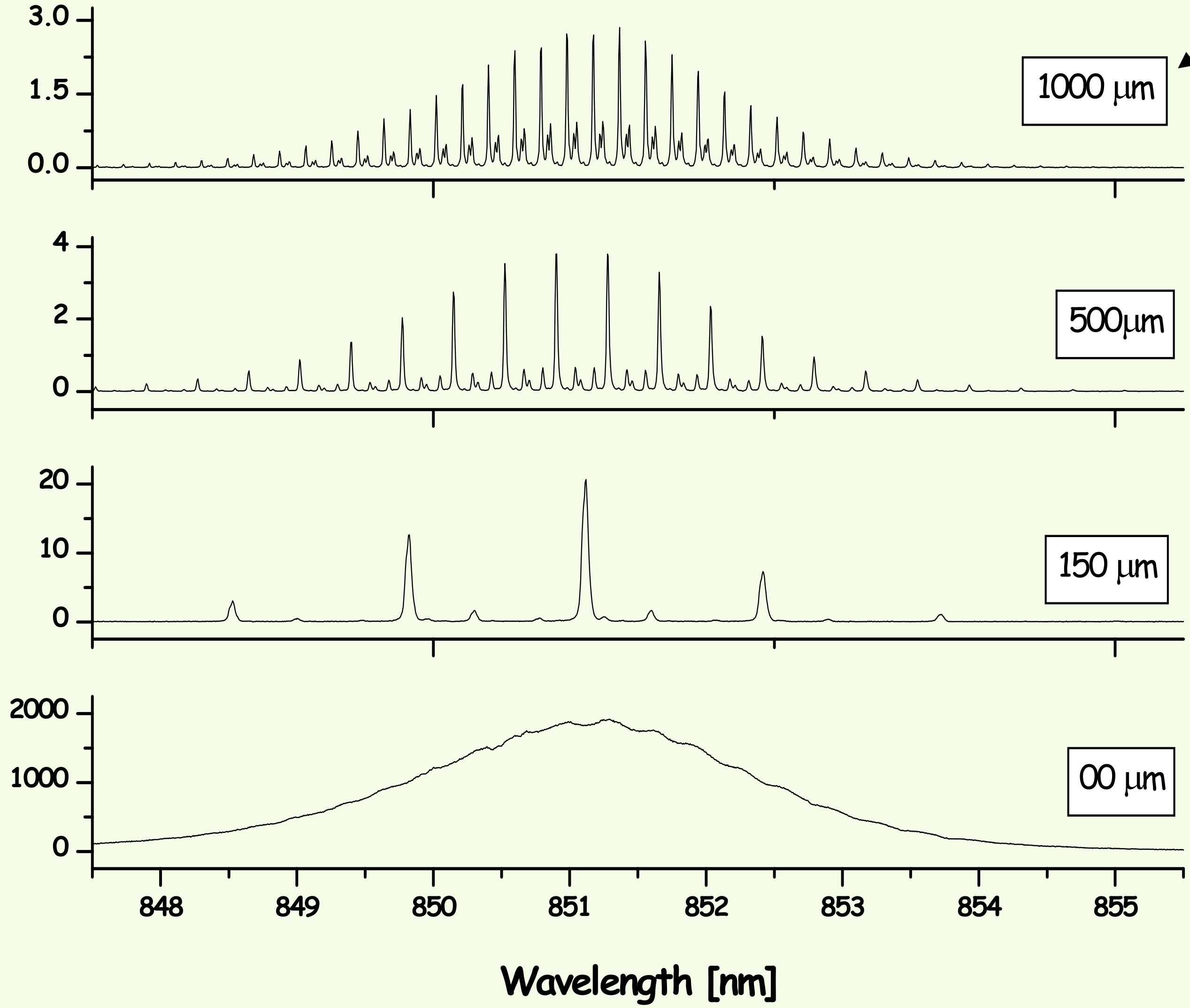


CEAS with a Mode-Locked laser



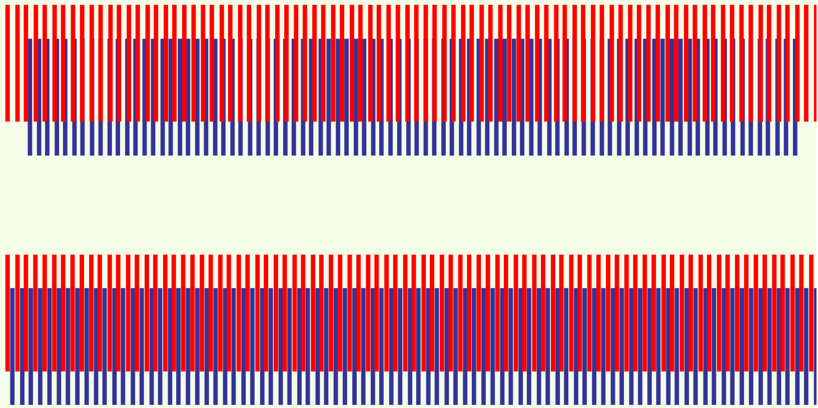
$M_{1,2}$: high reflectors around 800 nm

Beating between mode combs

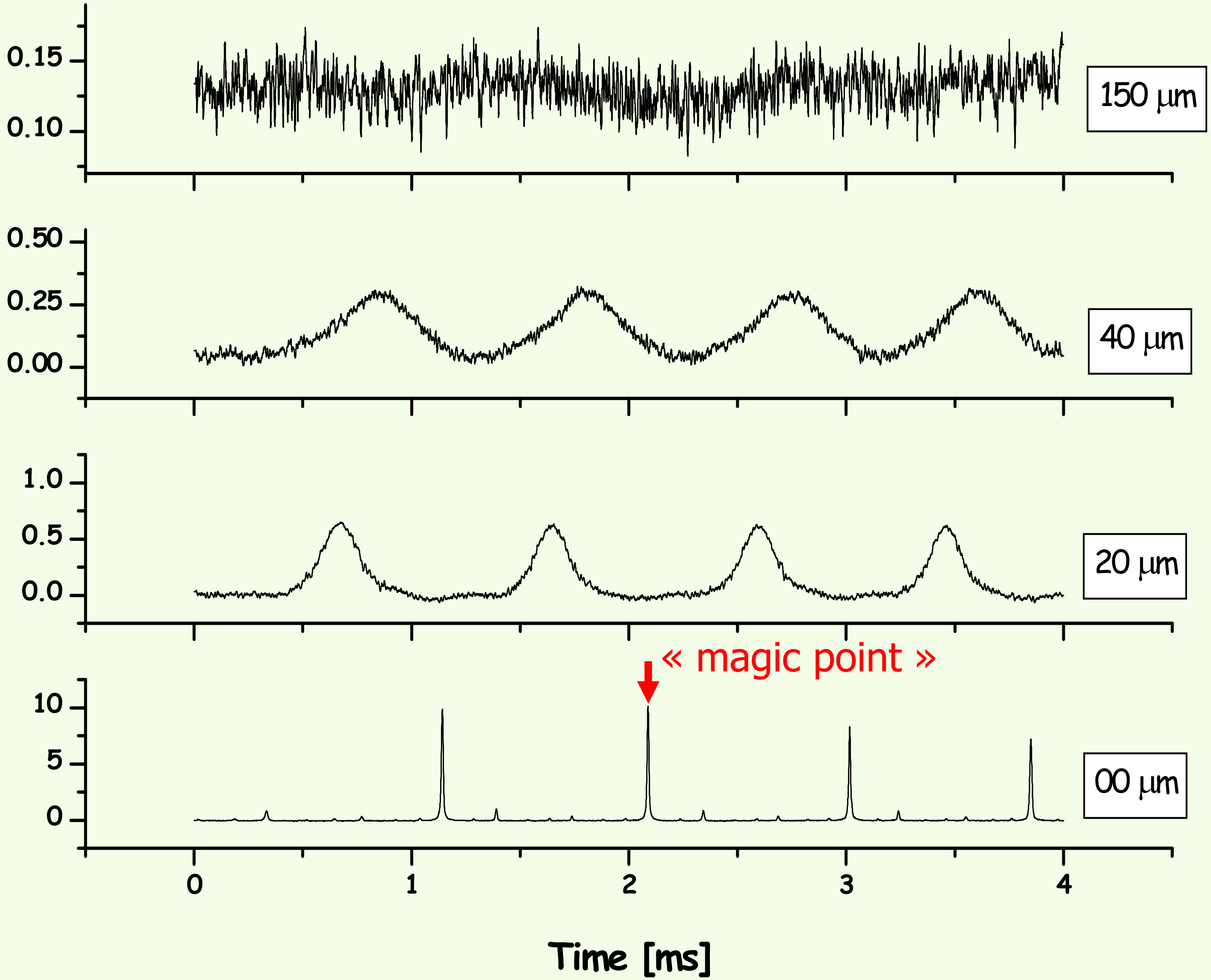


Transmitted spectra for different displacements of cavity length from the « magic point »

(smaller peaks are from transverse cavity modes)



Cavity transmission
(while slowly tuning its length)



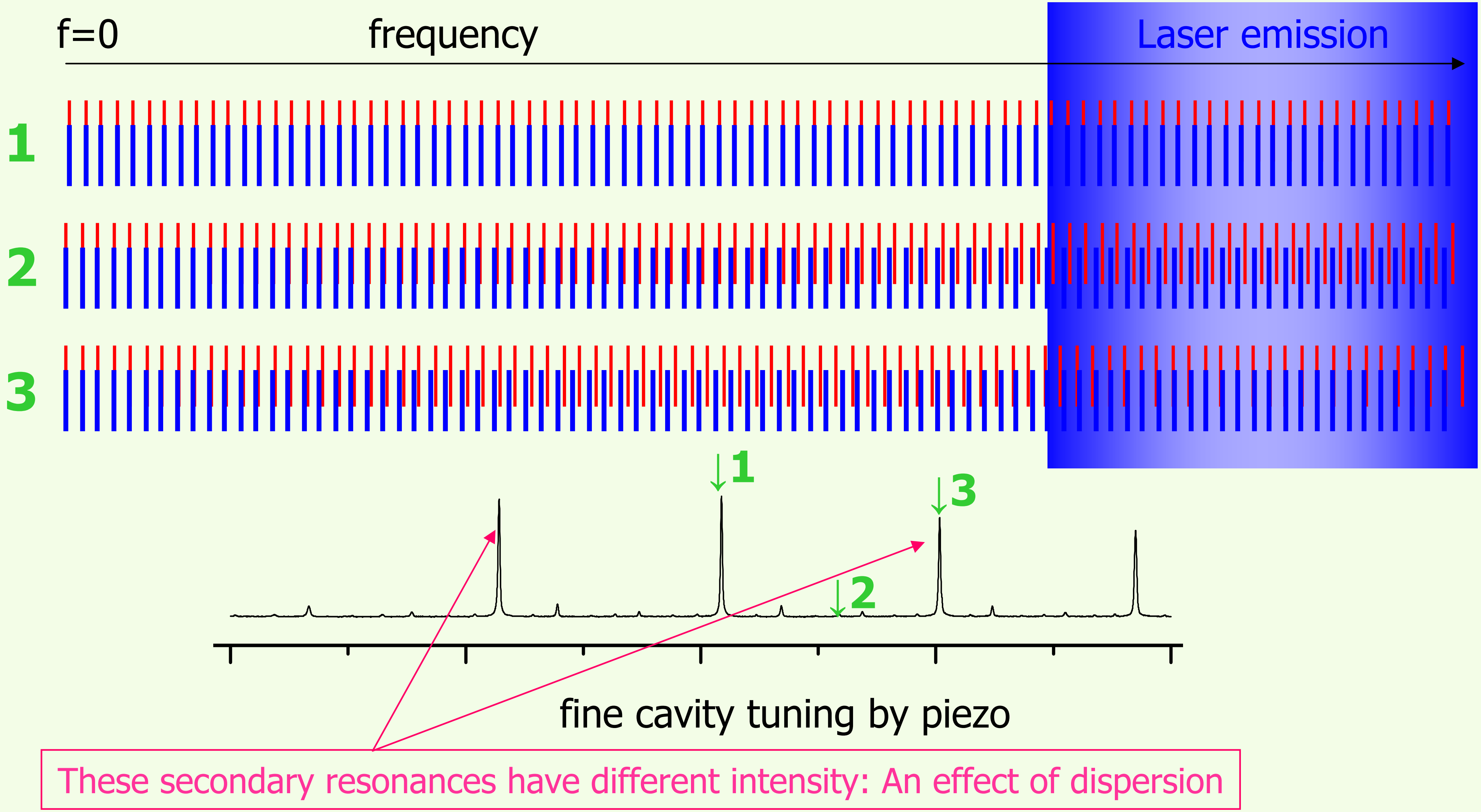
Cavity length tuning by a PZT

... distance from

« magic point »

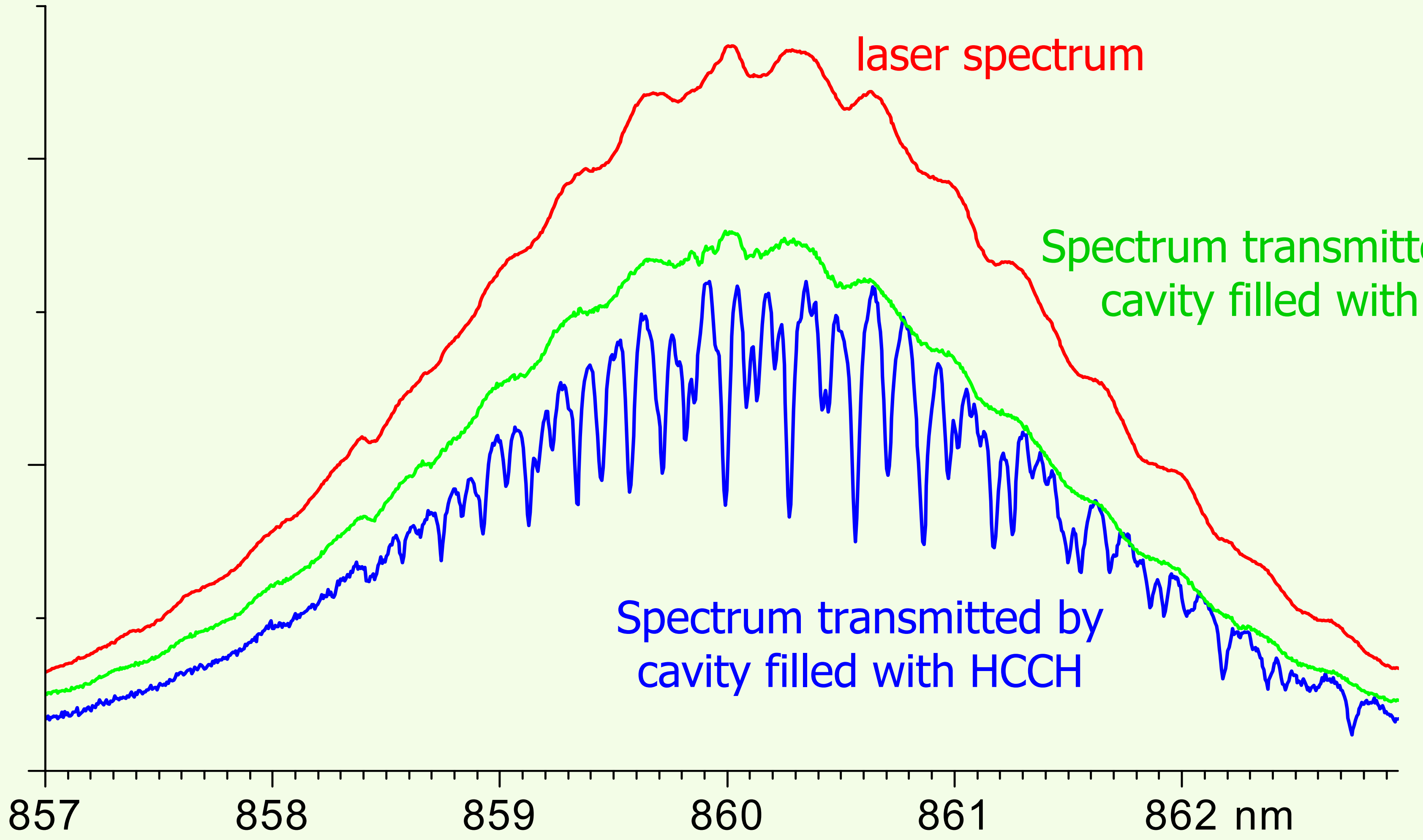
NOTE: When looking at these signals on a fast timescale, one can recover the pulse train again...

Why we see more than one comb resonances ?



To obtain spectra in transmission, one may either modulate the cavity around a resonance, or use a locking scheme

ML-CEAS : A first demonstration...

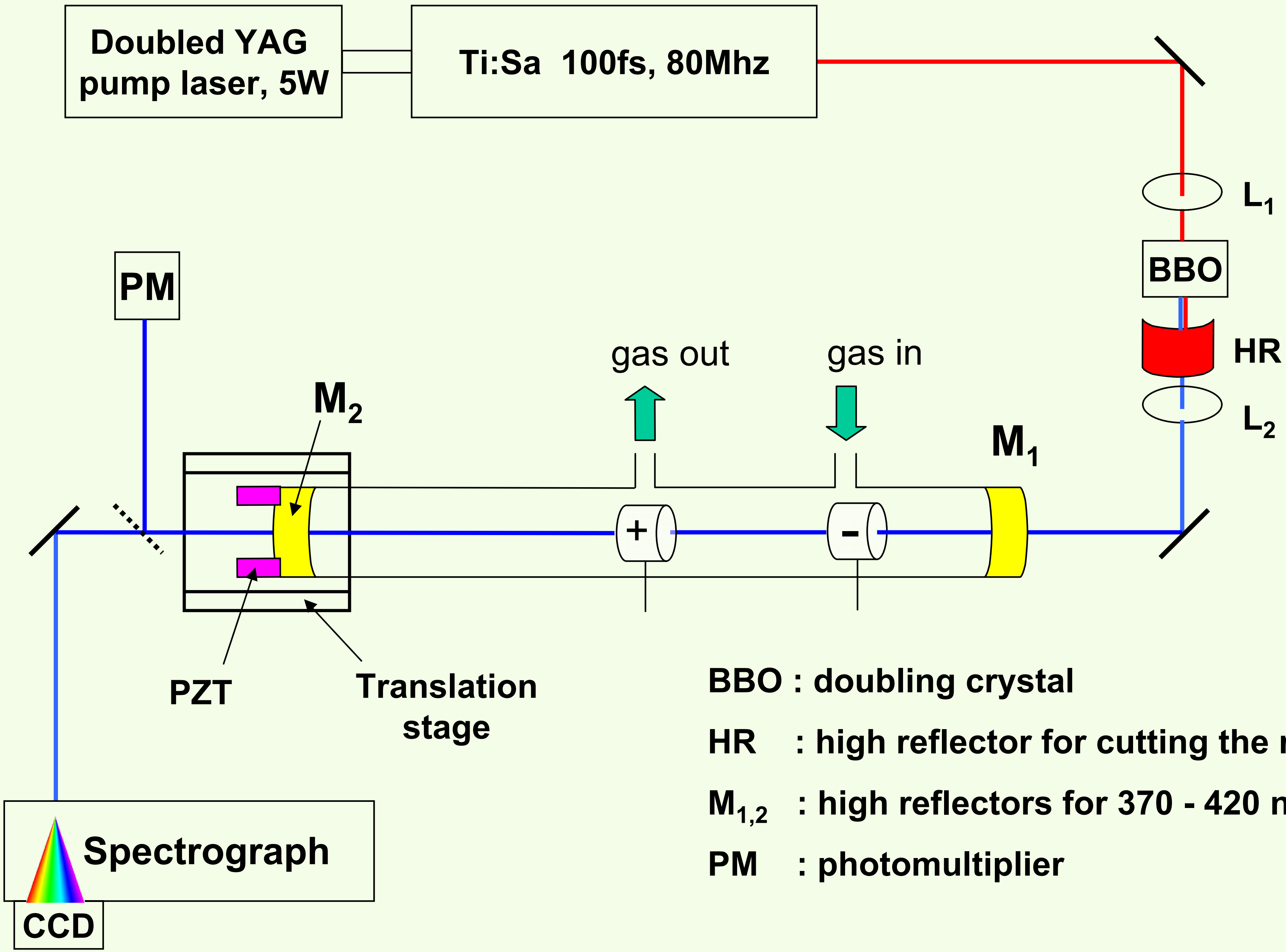


Effective absorption length F x L /π ~ 120 m

Acquisition time ~ 10 ms (cavity length was modulated…)

T. Gherman and D. Romanini, *Optics Express*, **10,** 1033 (2002).

Going to the UV



BBO : doubling crystal

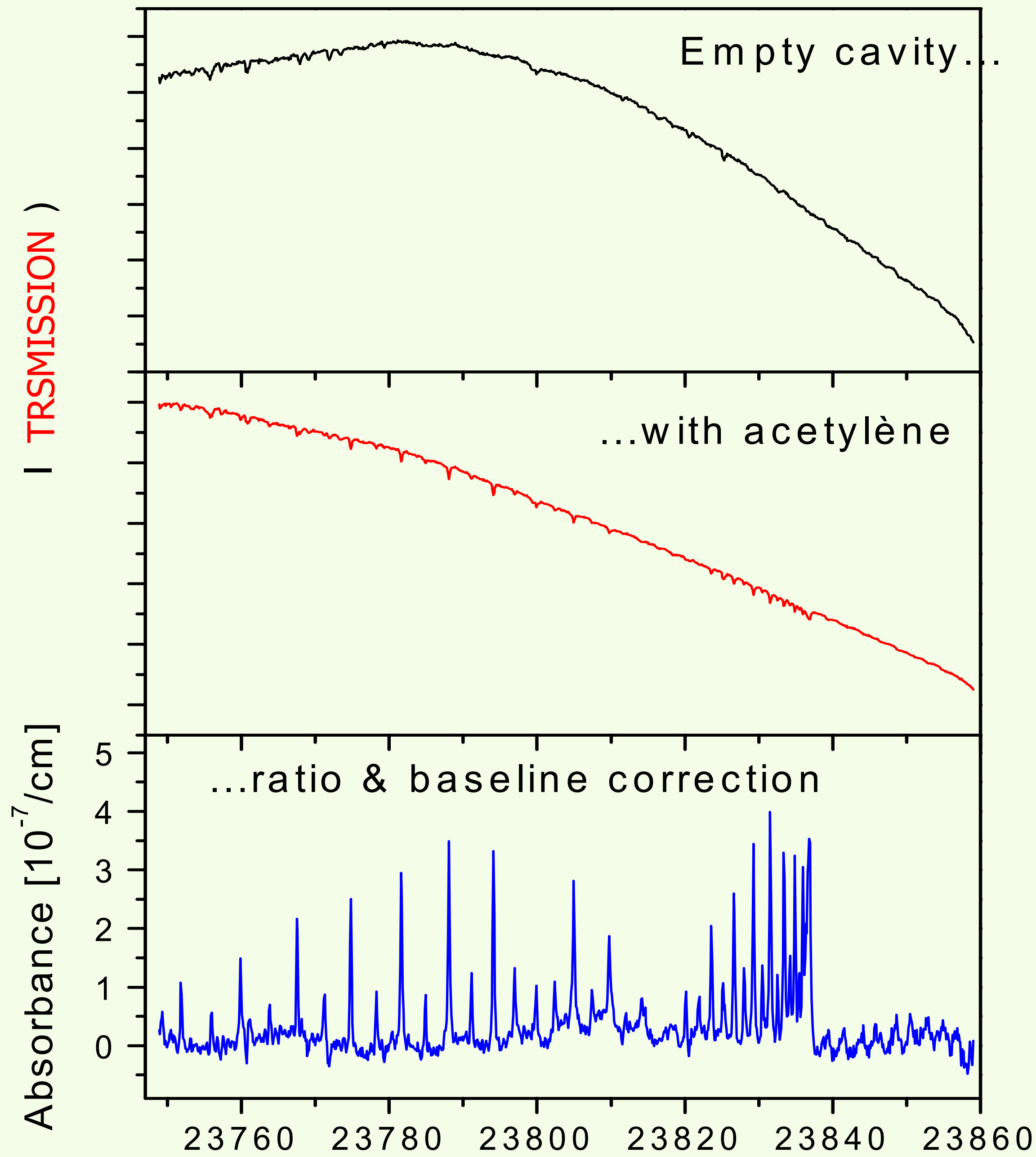
HR : high reflector for cutting the red

$M_{1,2}$: high reflectors for 370 - 420 nm

PM : photomultiplier

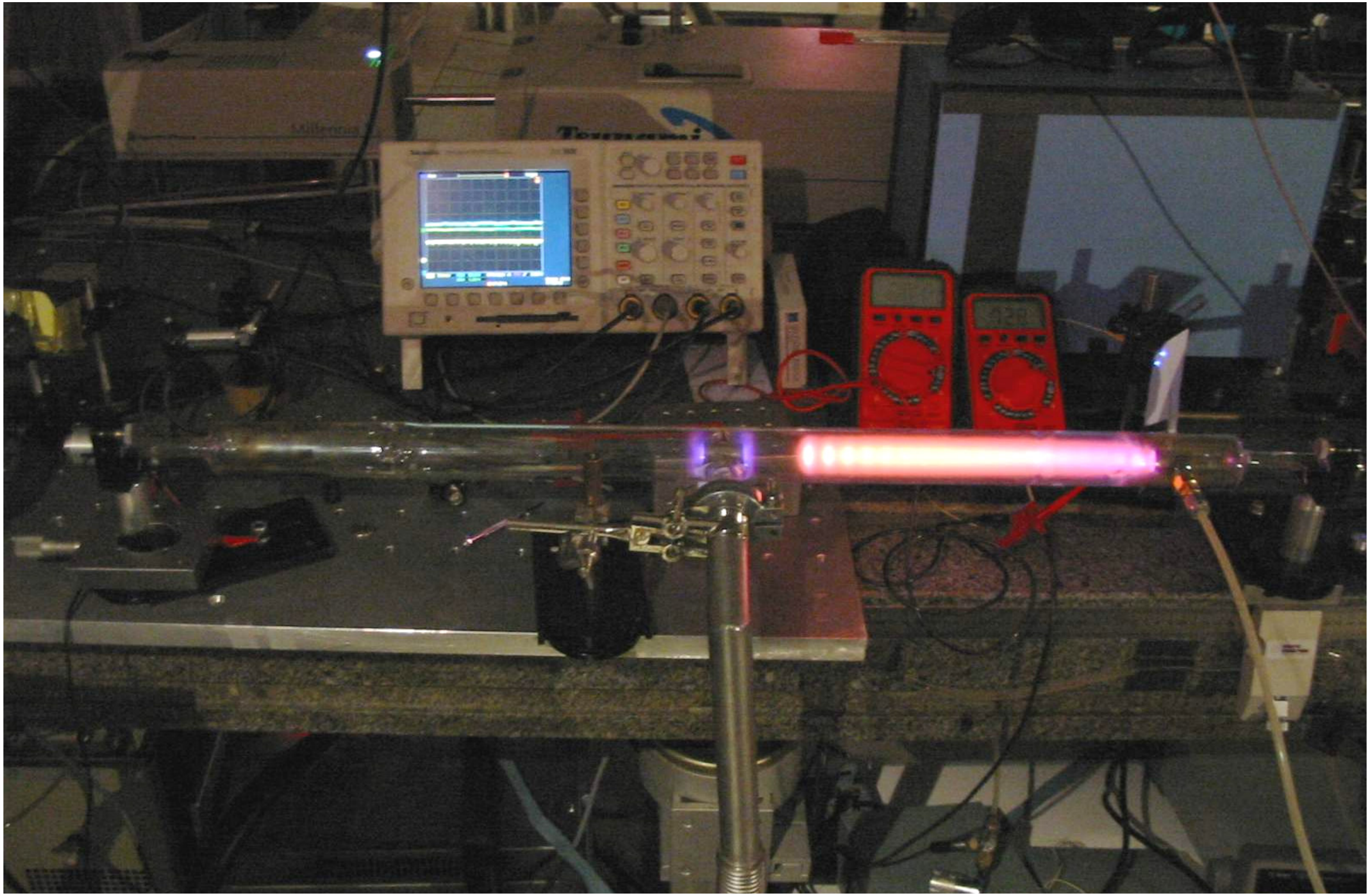
ML-CEAS : Application in the blue…

- Ti:Sa frequency doubled laser : Overtone transition of HCCH with 8 quanta of CH excitation (420 nm)
- Cavity Finesse ~ 3000
- Noise ~ 10^{-8} / cm
- Acquisition ~ 30 s, (using a basic frequency-locking scheme)

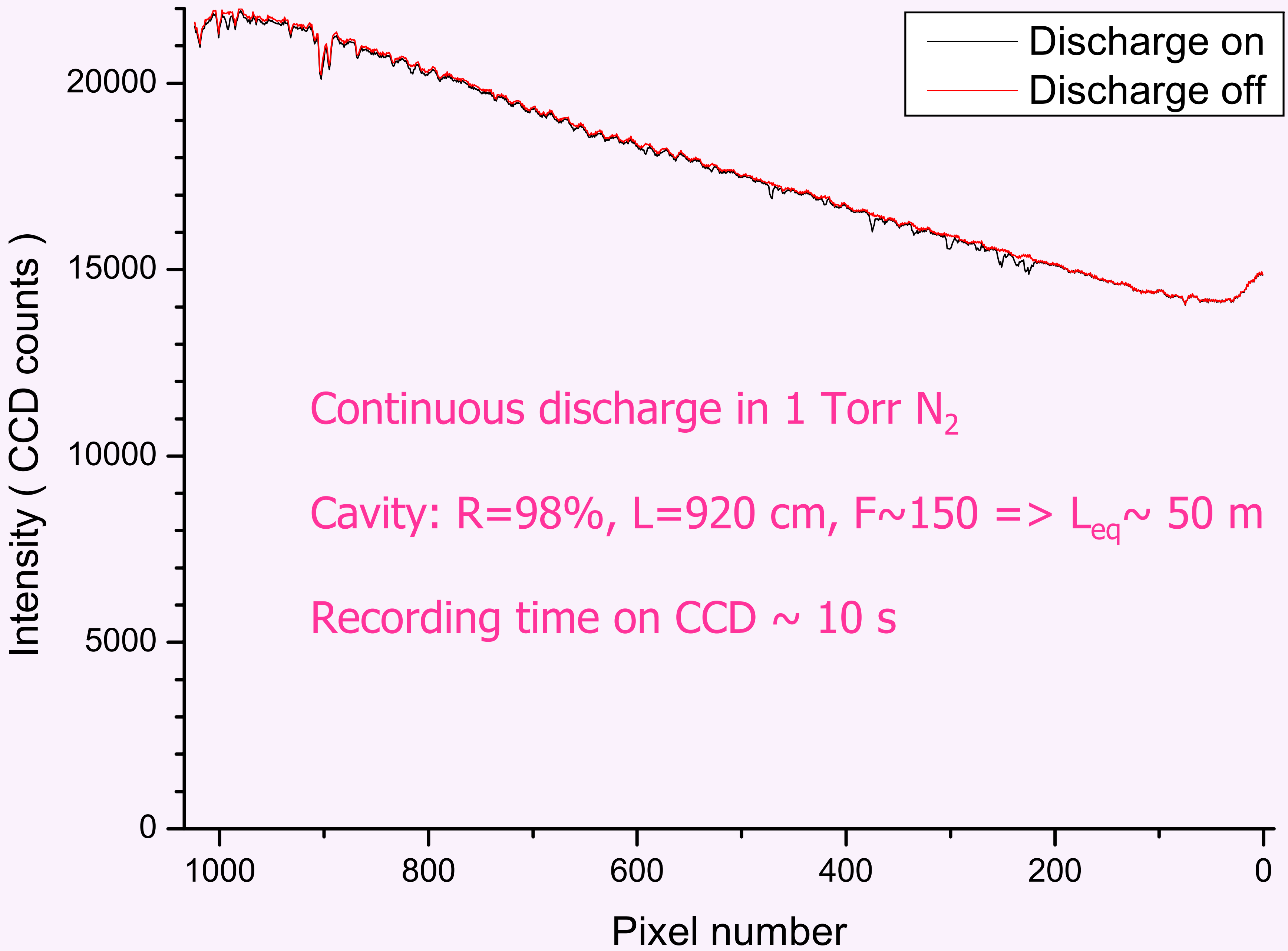


T. Gherman, S. Kassi, A. Campargue, D. Romanini
Chem. Phys. Letters **383** (2004) 353

ML-CEAS : N_2 discharge



Band 0-0 of N_2^+ ($B\ ^2\Sigma_u^+ - X\ ^2\Sigma_g^+$)

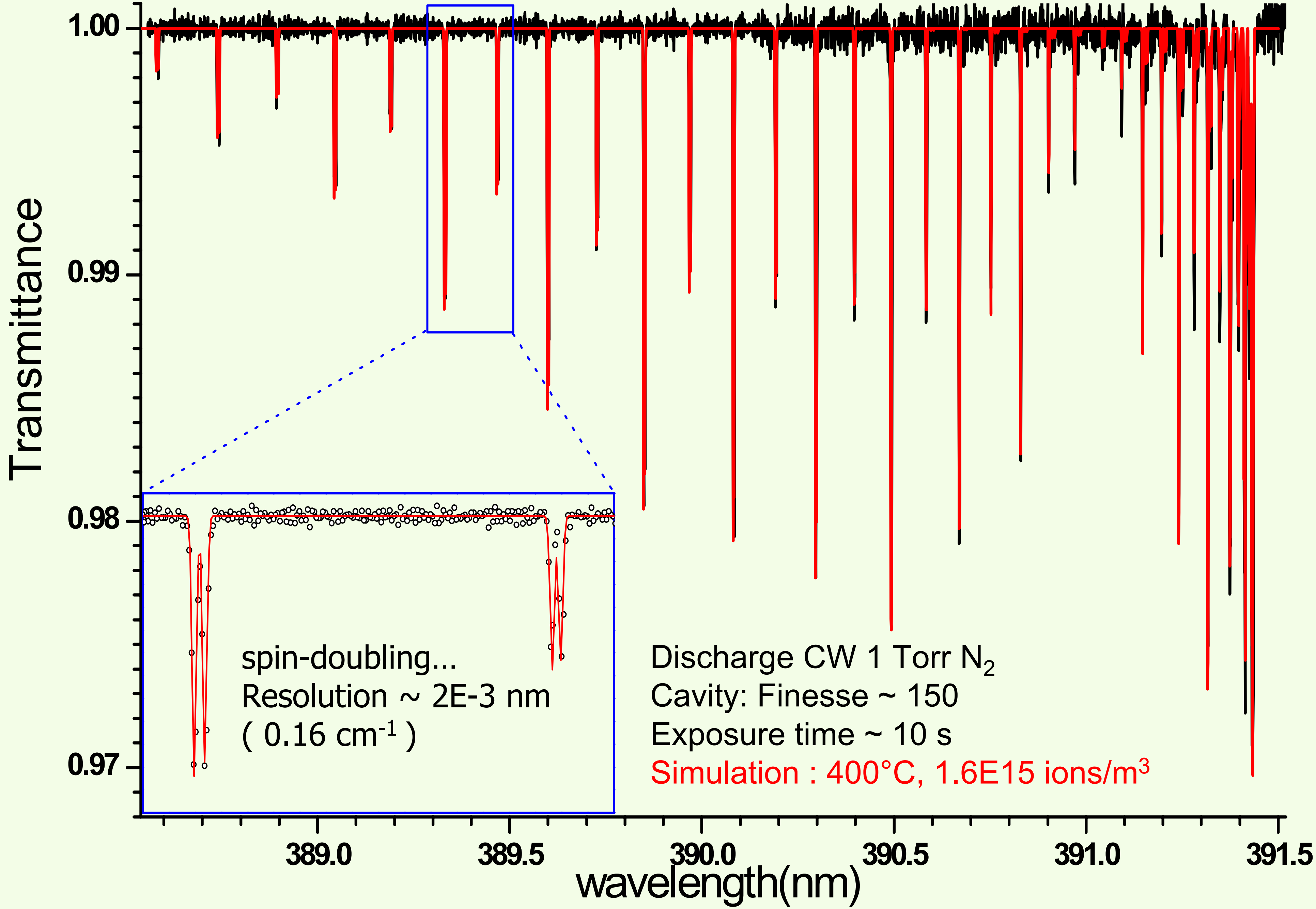


Continuous discharge in 1 Torr N_2

Cavity: R=98%, L=920 cm, F~150 => L_{eq}~ 50 m

Recording time on CCD ~ 10 s

Band 0-0 of N_2^+ $(B\ ^2\Sigma_u^+ - X\ ^2\Sigma_g^+)$



T. Gherman, E. Eslami, D. Romanini, J.-C. Vial, N. Sadeghi, *J Phys D* **37** (2004) 2408

Multiplex CEAS : ML-lasers versus Lamps (or LEDs)

Both sources are sufficiently smooth spectrally…

ML laser emission is highly coherent :

SPATIALLY (TEM00 mode) => Cavity mode-matching

… high cavity injection efficiency

… reproducible & accurate absorption measurements

TEMPORALLY (mode comb) => Cavity comb-locking

… further gain in injection

Lamps easily provide a larger emission spectrum :

Low resolution due to low spectral density and low cavity injection, and to the limited number of spectral elements on a CCD

Less expensive (except for electric consumption ?)

ML-CEAS, conclusions & perspectives

- **Robust, simple, multiplex** technique
- **Real-time, high sensitivity** ($2x10^{-9}/cm/Hz^{1/2}$)
- **Quantitative** (calibration of cavity finesse by ringdown)
- **=> UV** (efficient femtosecond pulse frequency upconversion)
- Small sample volume…
- **Frequency locking** of combs allows short acquisition times even with high finesse cavities…
- Effects of cavity **dispersion** (e.g. from mirrors) can be circumvented on relatively large spectral ranges (~>10nm) by using a small cavity modulation…