FREQUENCY METROLOGY WITH QUANTUM CASCADE LASERS

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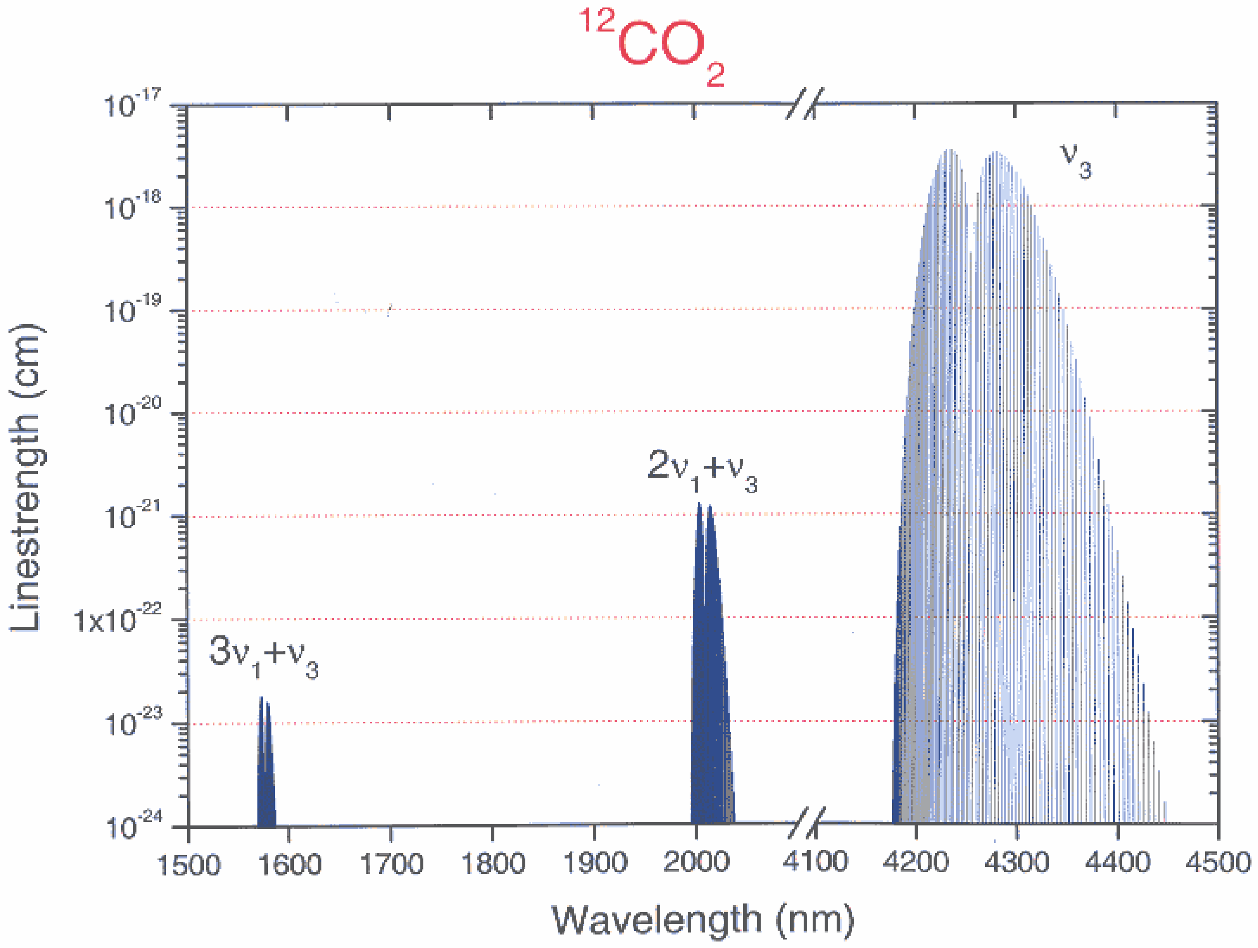
SCALING OF SENSITIVITY WITH WAVELENGTH

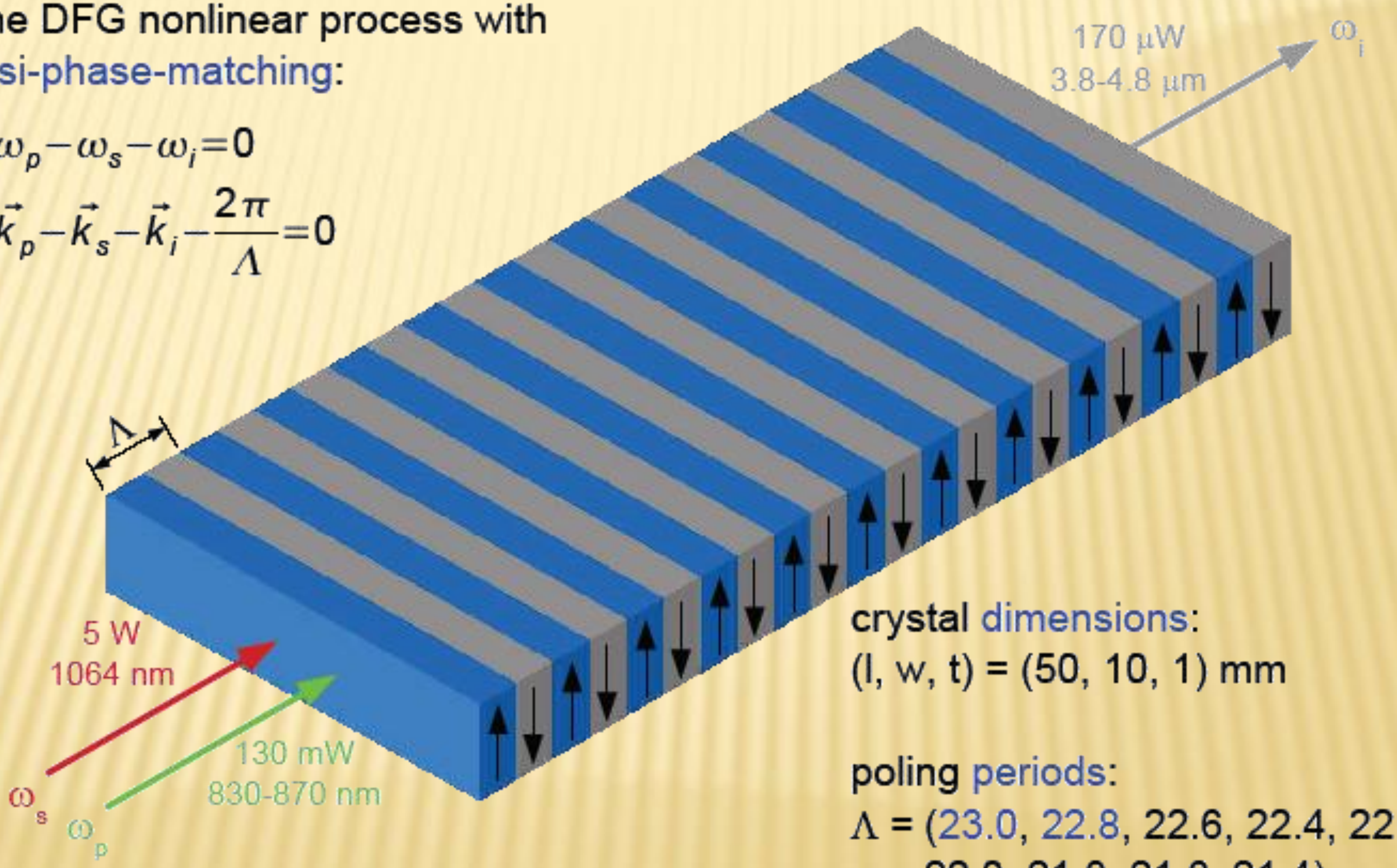


A POSSIBLE SOURCE:
DFG IN PP-CRYSTALS

Energy and momentum conservation in the DFG nonlinear process with quasi-phase-matching:

$$\omega_p - \omega_s - \omega_i = 0$$

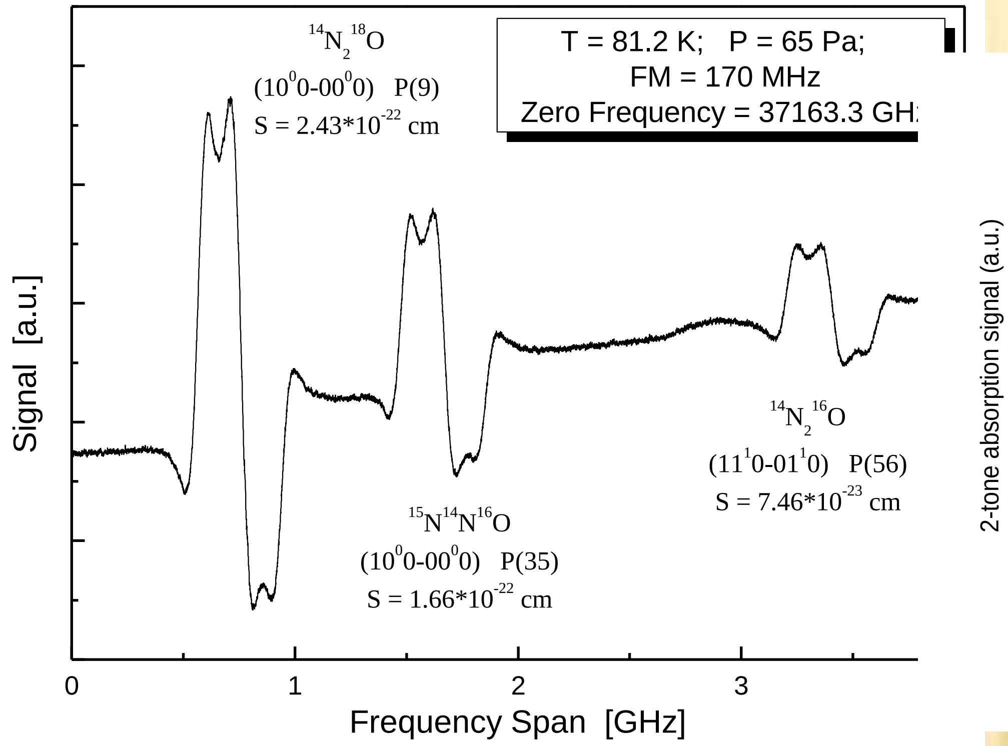
$$\vec{k}_p - \vec{k}_s - \vec{k}_i - \frac{2\pi}{\Lambda} = 0$$

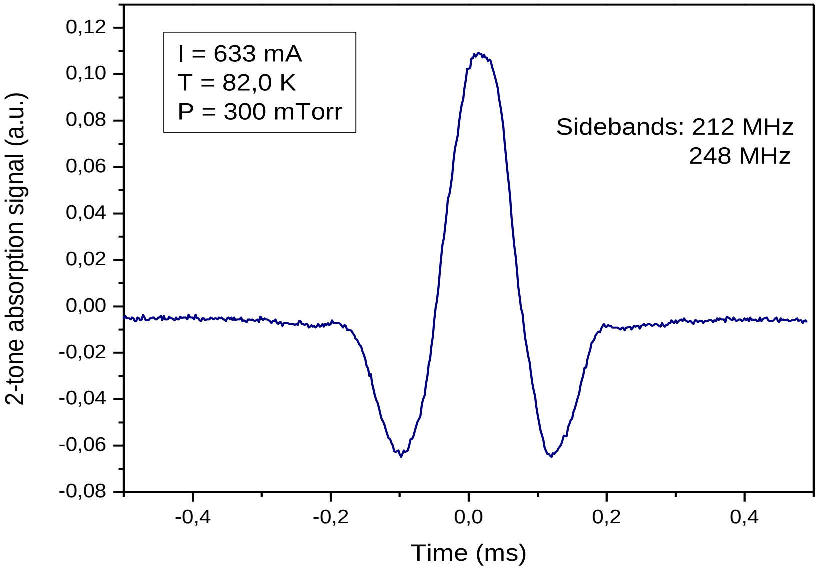


crystal dimensions:
(l, w, t) = (50, 10, 1) mm

poling periods:
Λ = (23.0, 22.8, 22.6, 22.4, 22.2,
22.0, 21.8, 21.6, 21.4) µm

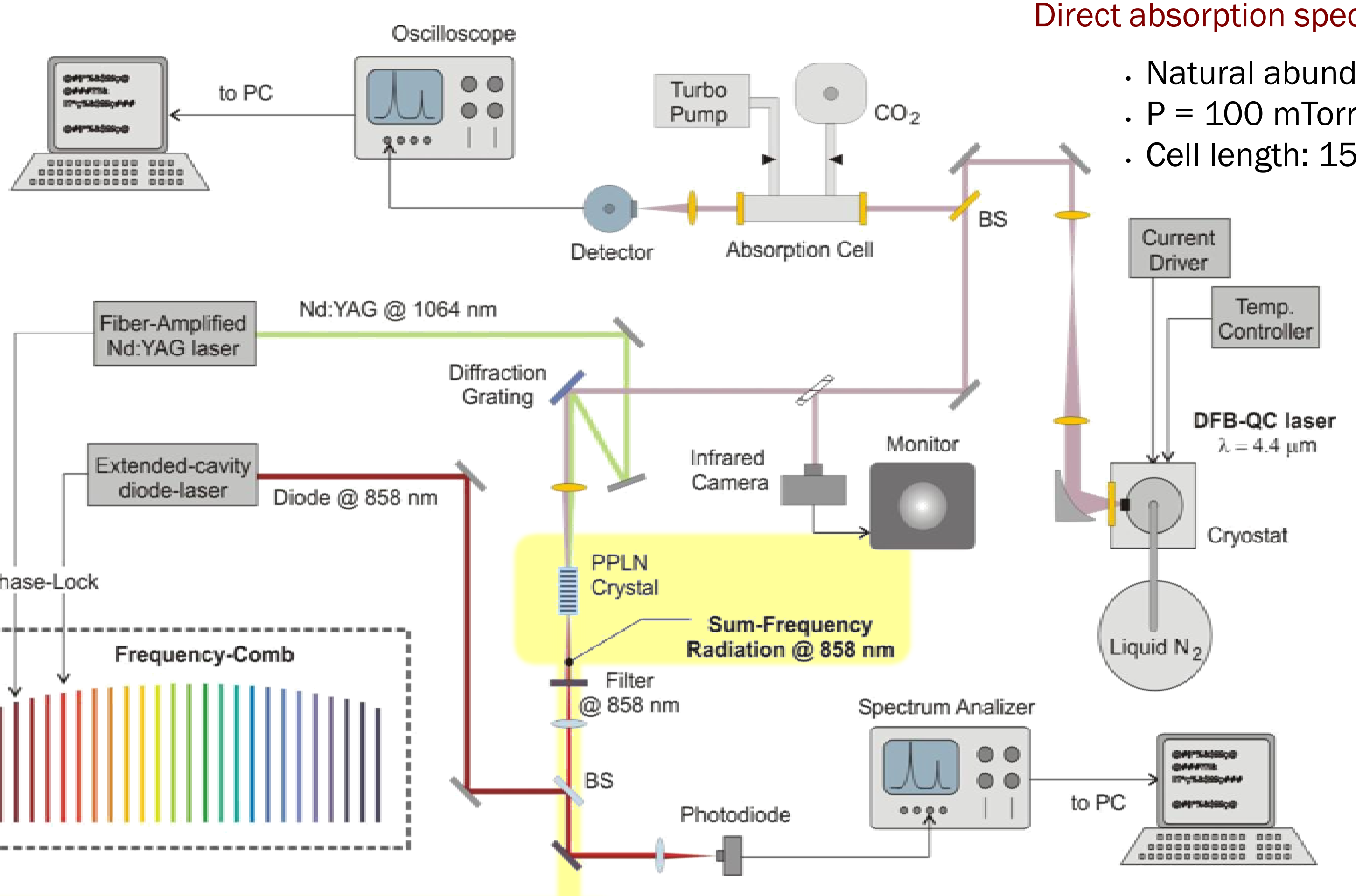
FM AND TWO-TONES MODULATION RESULTS





Borri et al., *Applied Physics B* 85, 223-229 (2006).

QCL-Comb Set-up



Direct absorption spectroscopy:

- Natural abundance CO_2 gas
- P = 100 mTorr
- Cell length: 15 cm

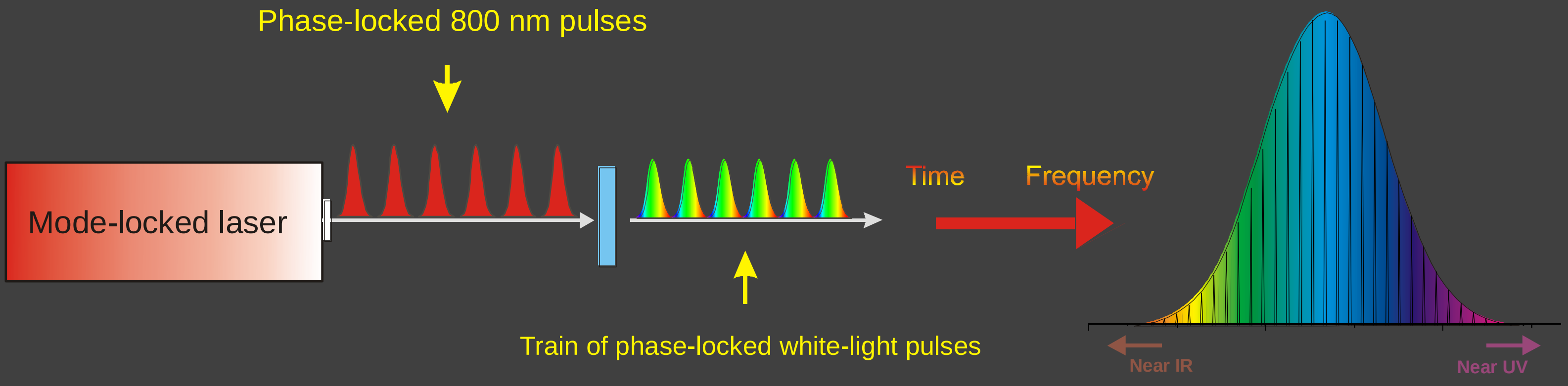
Sum frequency generation:

- Nd:YAG power on PPLN Crystal: 1.2 W
- QC Laser power on PPLN Crystal: 2.0 mW
- Generated power @ 858 nm: 7 μW

Beat-note detection:

- Power @ 858 nm: 7 μW
- Laser Diode power: 100 uW
- Beat-note amplitude: -35 dBm; SNR = 45 dB

THE OPTICAL FREQUENCY COMB SYNTHESIZER



Phase-locked 800 nm pulses

Mode-locked laser

Time

Frequency

Train of phase-locked white-light pulses

Near IR

Near UV

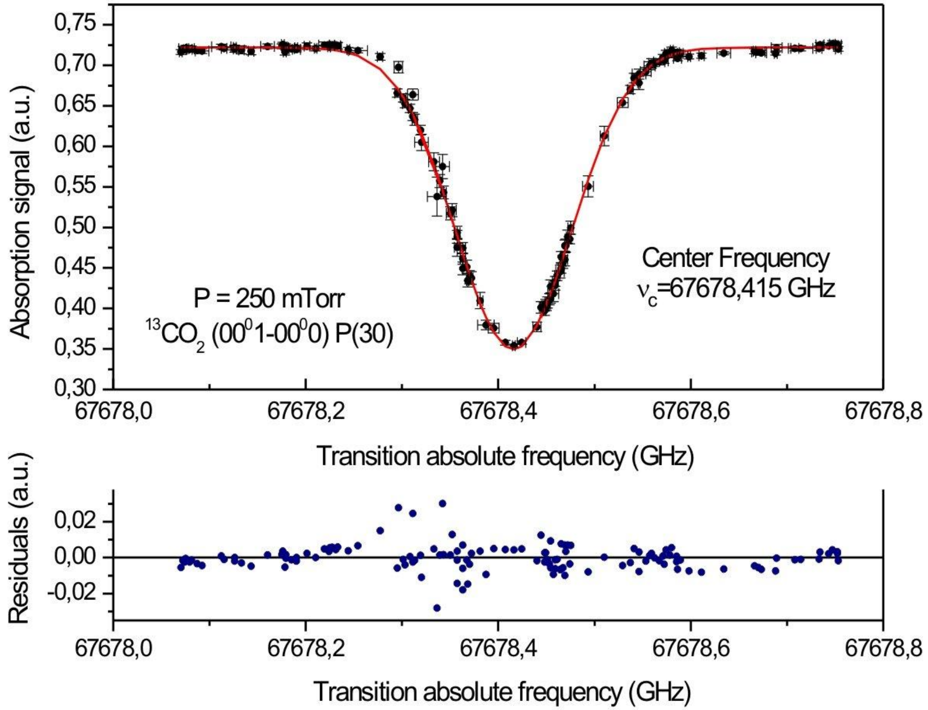
S. A. Diddams, D. J. Jones, J. Ye, S. T. Cundiff, J. L. Hall
J. K. Ranka, R. S. Windeler R. Holzwarth, T. Udem, and T. W. Hänsch

Physical Review Letters -- May 29, 2000 -- Volume 84, Issue 22, pp. 5102-5105

CO_2 line absolute frequency

Doppler Spectroscopy

$^{13}CO_2$ $(00^01\text{-}00^00)$ P(30)

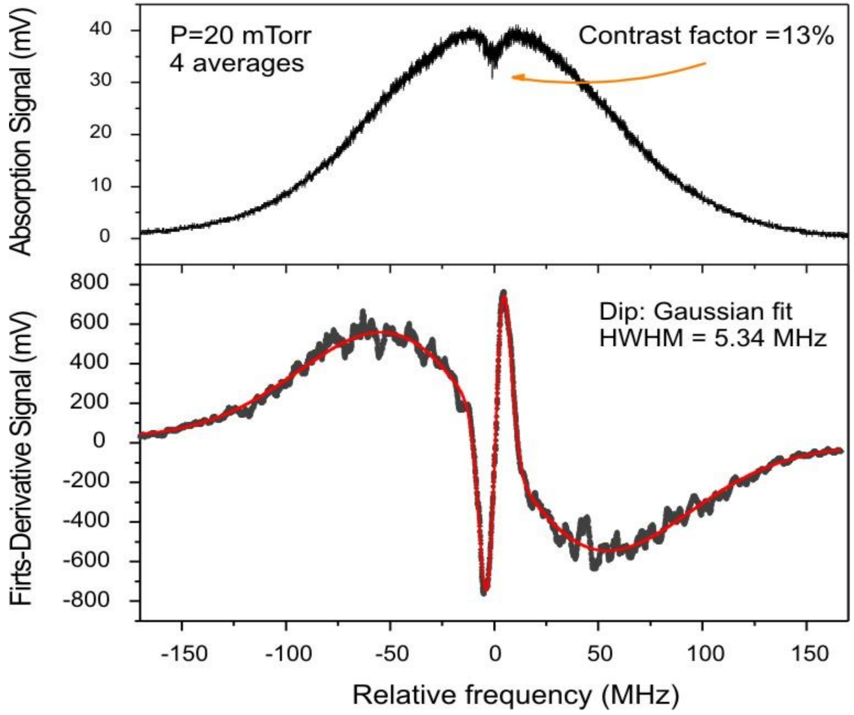


N$_C$ = 67 678 415 ± 2 MHZ

S. Bartalini et al., *Opt. Lett.* 32, 988 (2007)

Sub-Doppler Spectroscopy

$^{12}CO_2$ $(01^11\text{-}01^10)$ P(30)

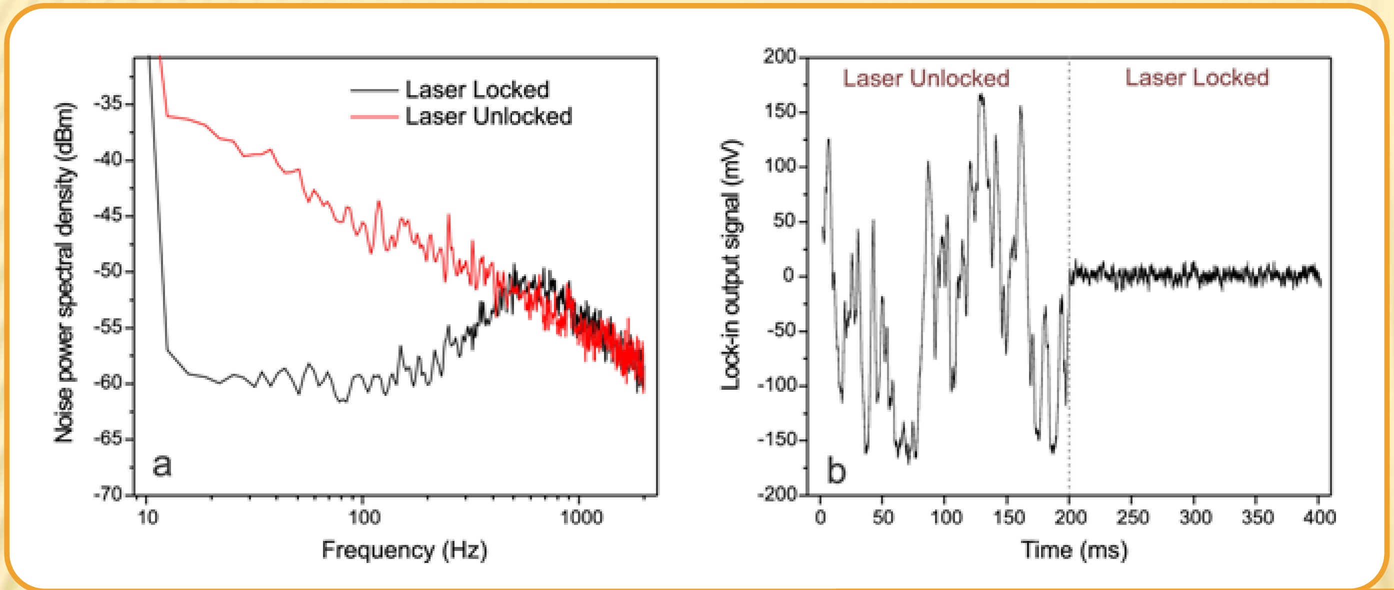


kHz-level precision on dip-center frequency determination

S. Borri et al., *Opt. Express* 16, 11645 (2008)

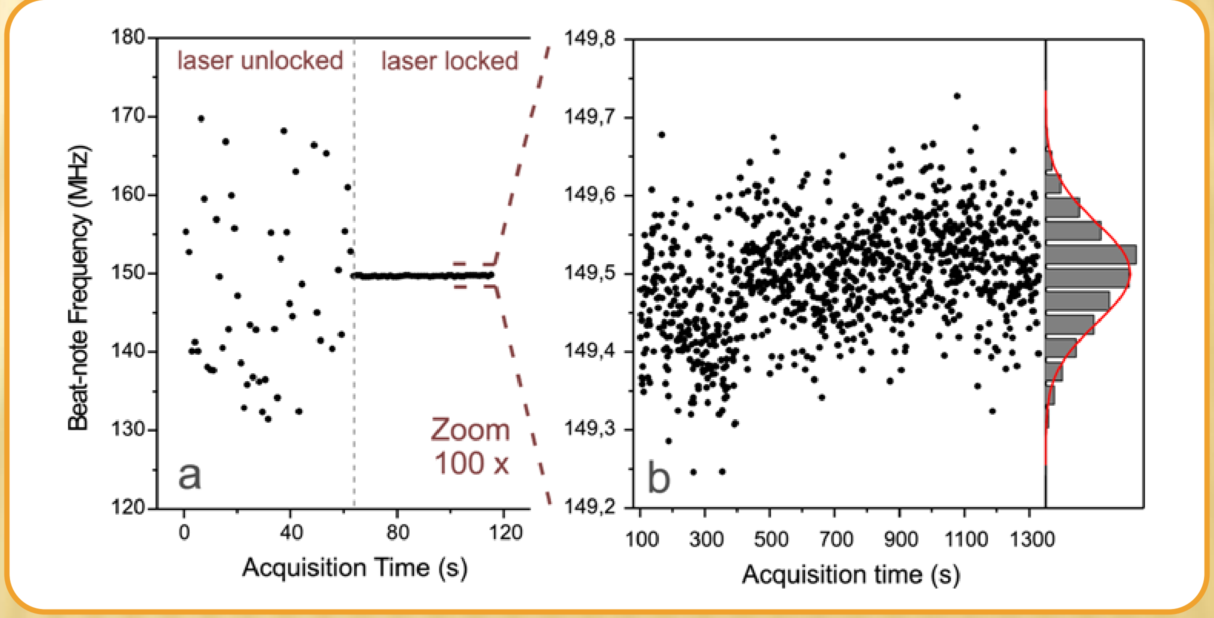
Lamb-dip-locked quantum cascade laser for comb-referenced IR absolute frequency measurements - 2/3

Frequency locking on the first derivative signal

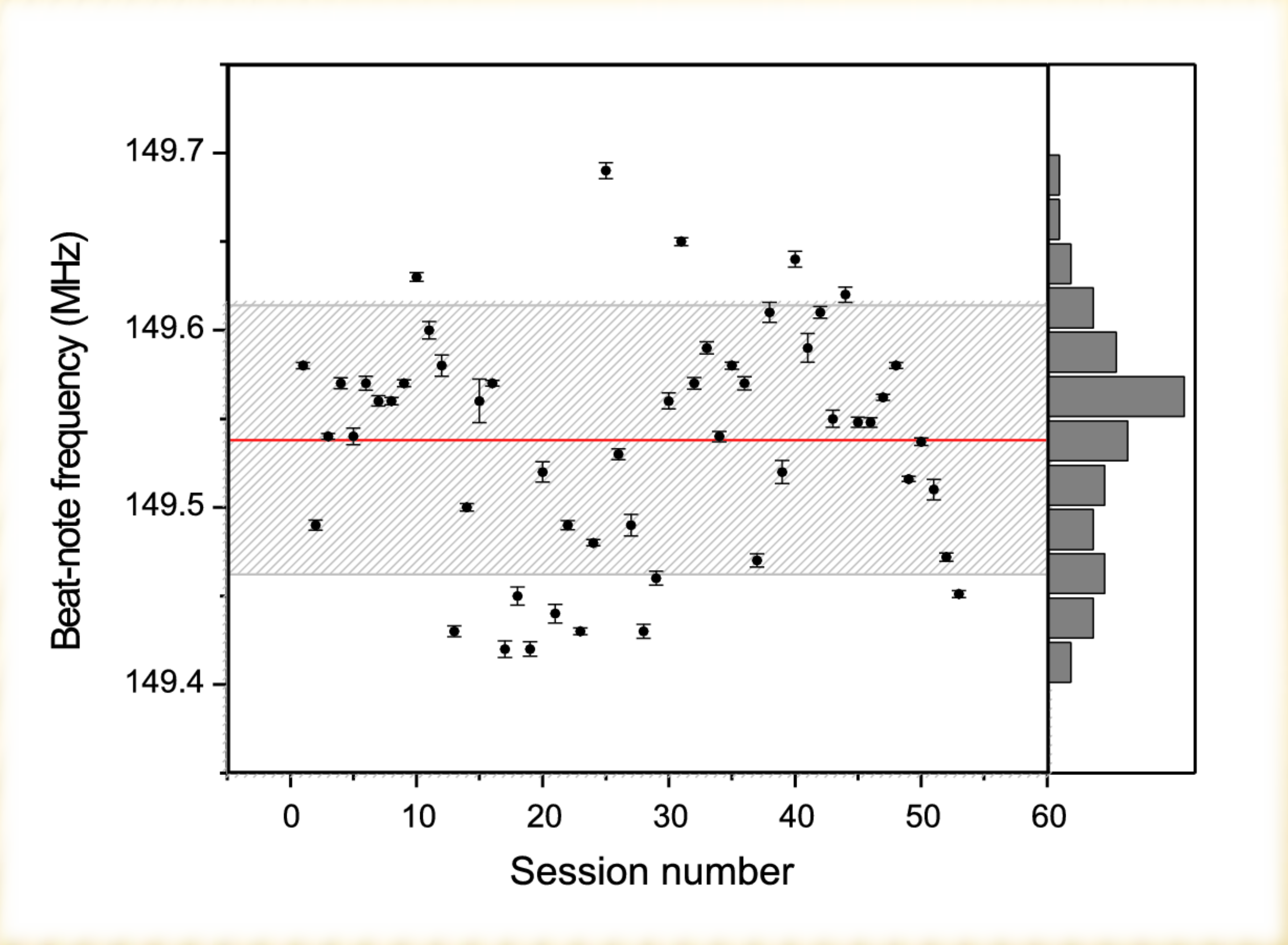


- 25 dB noise reduction at low frequencies
- Locking bandwidth limited to a few hundred Hz due to the lock-in time constant
- The wide low-frequency oscillations are cut-off

- Unlocked laser: frequency oscillations of tens of MHz between each of the 1-second-long measurements
- Locked laser: the frequency fluctuations are greatly reduced to hundreds of kHz
- The precision of each data-set (figure b) can be assumed to be a few kHz (standard deviation of the mean)



Lamb-dip-locked quantum cascade laser for comb-referenced IR absolute frequency measurements - 3/3



- Final accuracy of 75 kHz (safe estimation) limited by systematics
- We expect an improvement of a factor 100 by improving the laser stability and the locking loop

Lamb-dip-locked quantum cascade laser for comb-referenced IR absolute frequency measurements

S. Borri, S. Bartalini, I. Galli, P. Cancio, G. Giusfredi, D. Mazzotti, A. Castrillo, L. Gianfrani and P. De Natale

Optics Express **16**, 11637 (2008)

Open Problems

Intensity oscillations:

- Mechanical instabilites affect the aligment of the laser beam.
- Strong absorption in air attenuates the laser power. Moreover the air turbolence along the optical path leads to random amplitude oscillations of the signal.
- Optical interference fringes introduce systematic errors in determining the locking point.

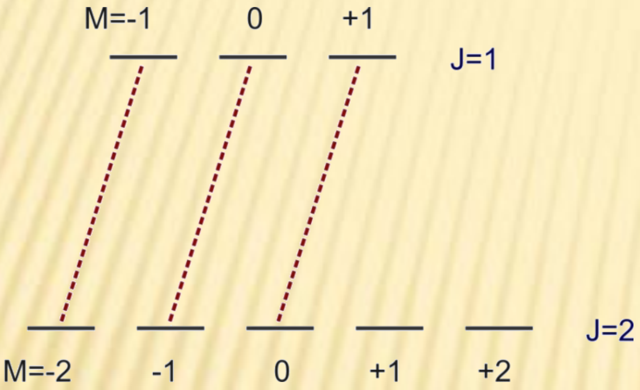
Locking loop related problems:

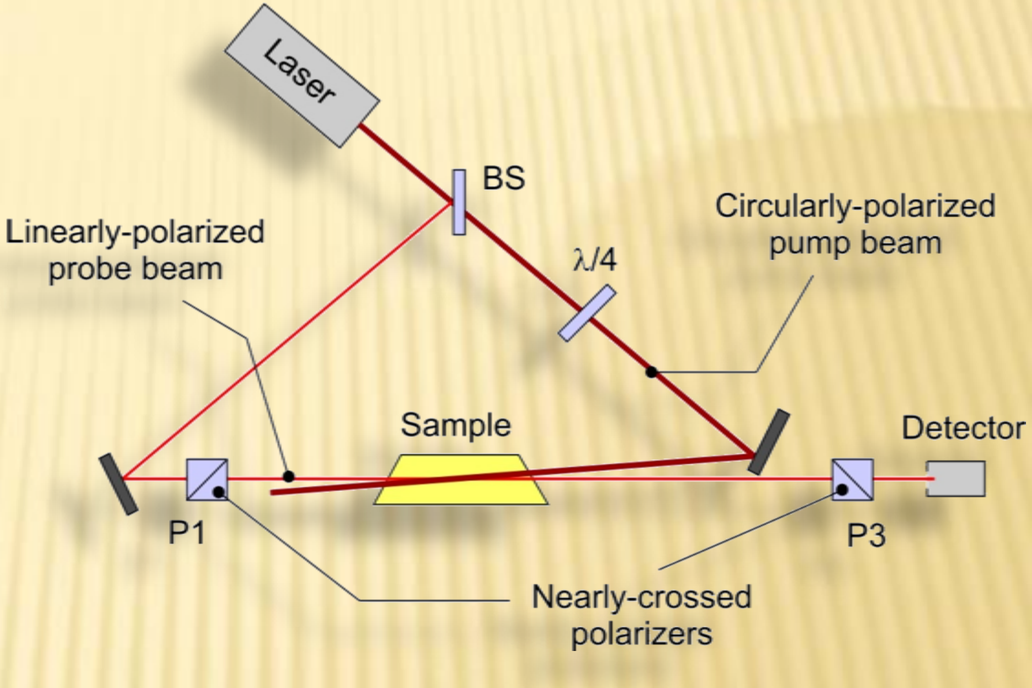
- The modulation of the laser frequency is limited by the actual bandwidth of the current driver modulation input (about 150 kHz).
- The locking bandwidth is actually limited by the time constant of the lock-in amplifier used for the demodulation.
- We have intentionally avoided a direct HF signal coupling to the QC laser, in order to minimize the risk of electrostatic shock damages.

Doppler-Free Laser Polarization Spectroscopy

A very high-sensitivity technique largely used in the visible and near-IR

- need of good polarizing wave-plates
- need of excellent polarizers and polarizing beam splitters





Because of the selection rules, **not all of the M sublevels are pumped**.

The pumping process produces an unequal saturation and, with it, **a nonuniform population of the M sublevels**, which is equivalent to an anisotropic distribution for the orientations of the angular momentum vector J.

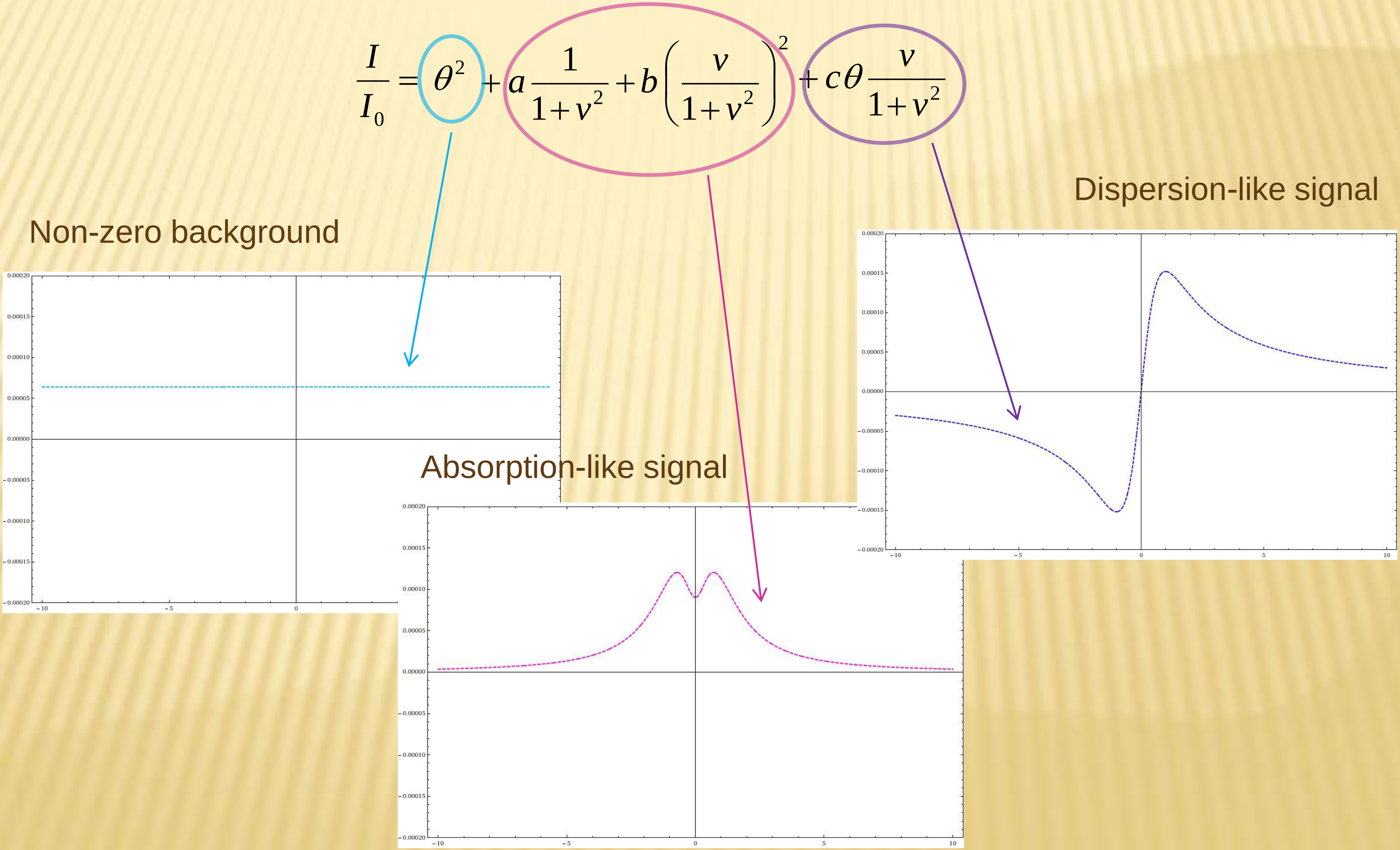
Such an anisotropic **sample becomes birefringent for the incident linearly polarized probe beam**. As a consequence, its plane of polarization is slightly rotated after the interaction with the anisotropic sample.

→ *C. Wieman and T. W. Hänsch*, Phys. Rev. Lett. **36**, 1170 (1976)

Polarization spectroscopy

Signal variation with the angle θ of the polarizer P3.

We assume θ=0 as the angle corresponding to P3 crossed with P1.



$$\frac{I}{I_0} = \theta^2 + a\frac{1}{1+v^2} + b\left(\frac{v}{1+v^2}\right)^2 + c\theta\frac{v}{1+v^2}$$

Non-zero background

Absorption-like signal

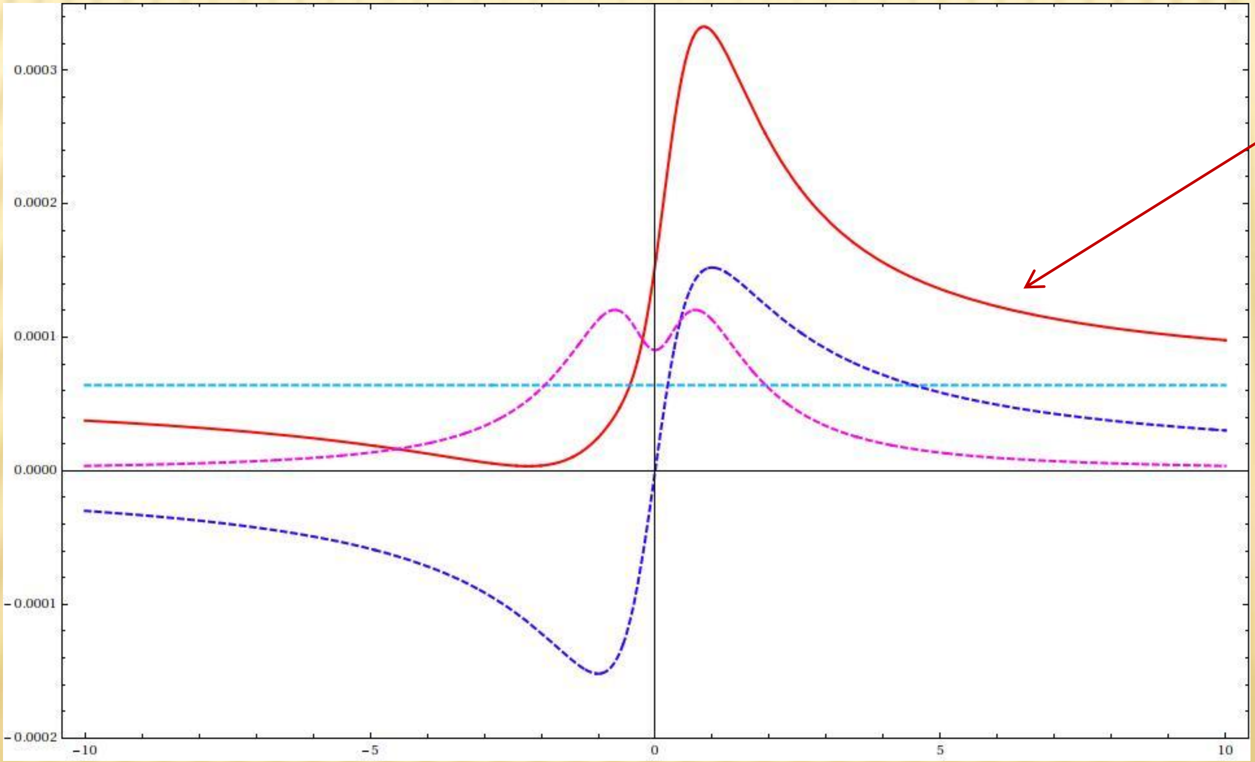
Dispersion-like signal

Polarization spectroscopy

Signal variation with the angle θ of the polarizer P3.

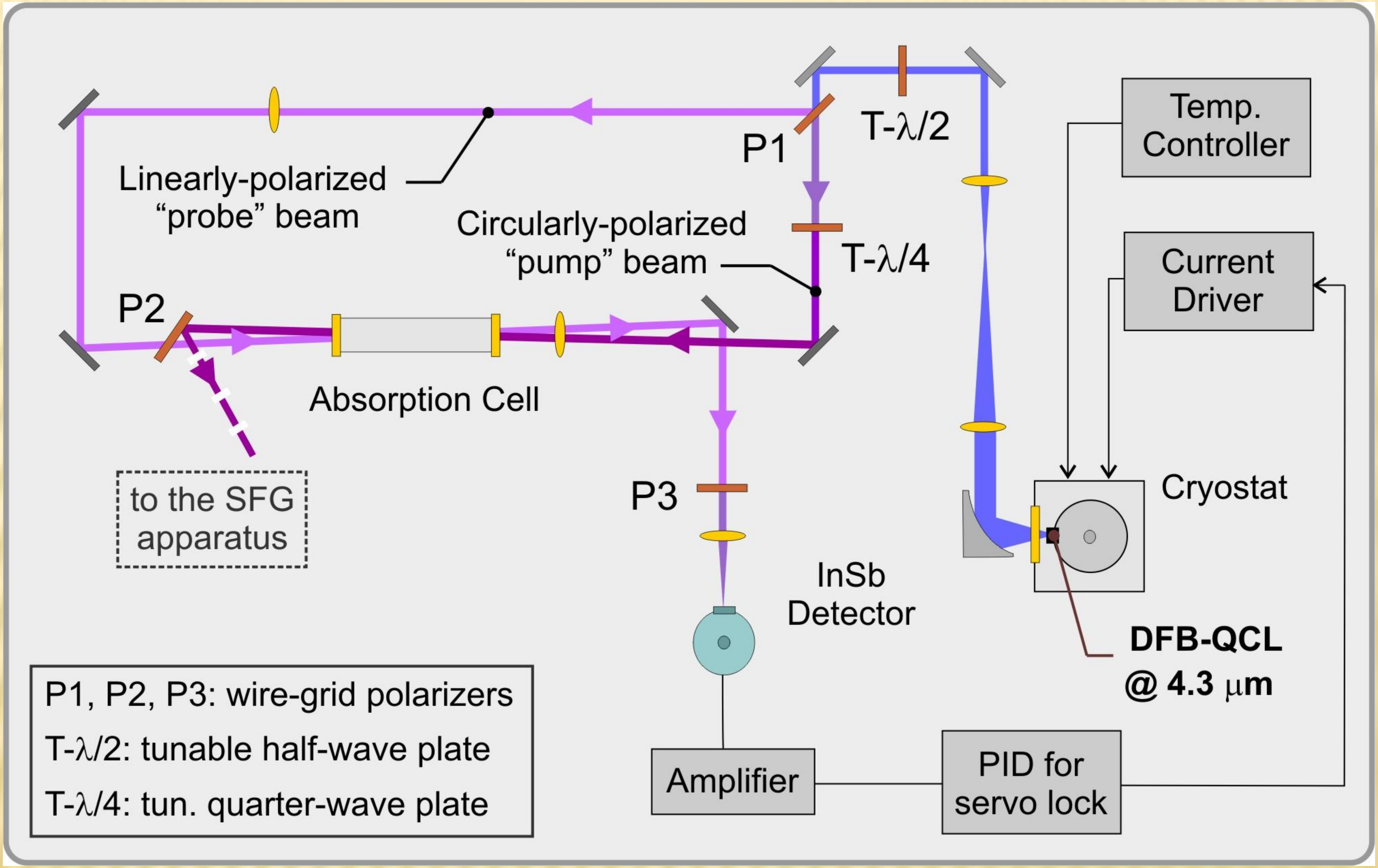
We assume θ=0 as the angle corresponding to P3 crossed with P1.

$$\frac{I}{I_0} = \theta^2 + a\frac{1}{1+v^2} + b\left(\frac{v}{1+v^2}\right)^2 + c\theta\frac{v}{1+v^2}$$



Polarization overall signal

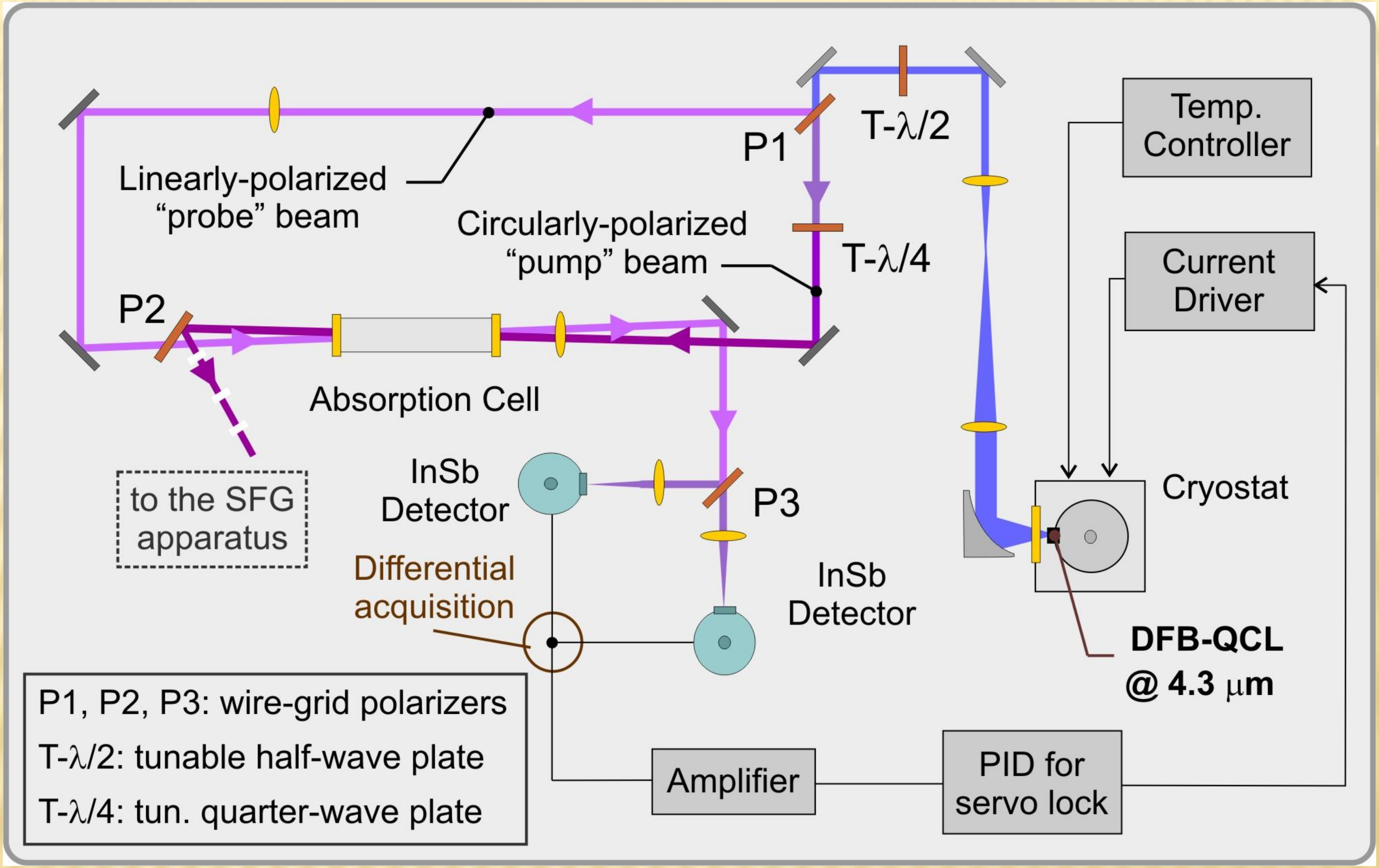
Polarization spectroscopy



P1, P2, P3: wire-grid polarizers
T-λ/2: tunable half-wave plate
T-λ/4: tun. quarter-wave plate

Polarization spectroscopy is ideal for locking the laser with no need of external modulation. In the basic setup, P1 and P3 are nearly crossed polarizers.

Polarization spectroscopy: towards a differential acquisition setup



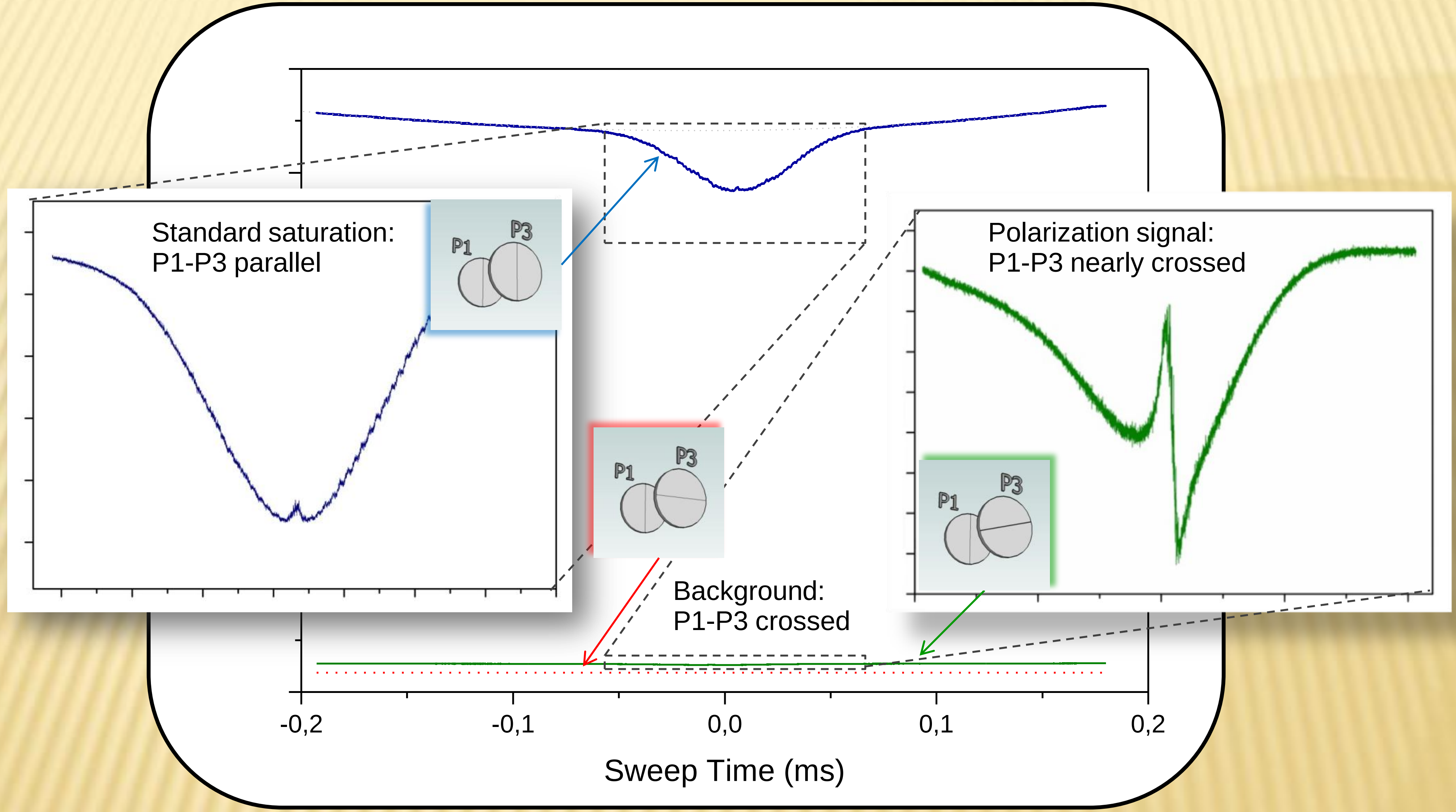
P1, P2, P3: wire-grid polarizers

T-λ/2: tunable half-wave plate

T-λ/4: tun. quarter-wave plate

From a differential detection we expect a great improvement of both the signal-to-noise ratio of the polarization signal and its symmetry.

Polarization spectroscopy



“Pure” saturation spectrum: in absence of the crossed polarizer on the probe beam (P3, in front of the detector) the Lamb dip is recorded

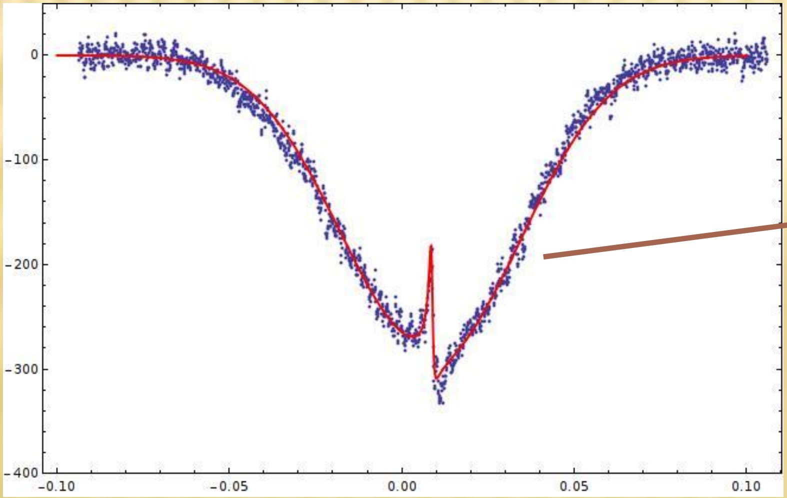
Polarization spectrum: the crossed polarizer on the probe beam allows to detect the polarization effect. The dip is greatly enhanced with respect to the Doppler profile. This signal can be used for locking the laser with no need of modulations

Polarization spectroscopy

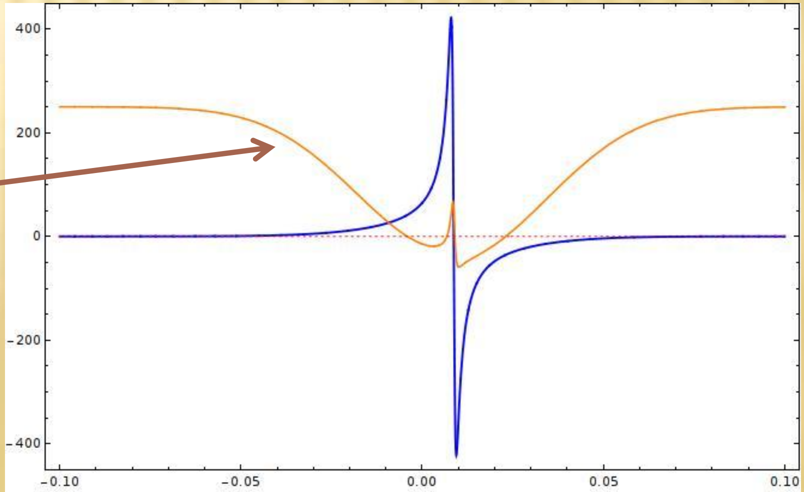
Towards a higher sensitivity detection:

- ✓ Improved stabilization of the QCL and mechanical isolation of the optical table
- ✓ Full coverage of the apparatus to stabilize absorption in air
- ✓ Nitrogen flux to minimize absorption in air (from 75% to 10%)
- ➢ Setting up a differential acquisition scheme (in progress)
- ➢ Setting up a high-bandwidth coupling of the correction signal to the laser (already designed)

Below: good agreement between the experimental data (blue dots) and theoretical polarization curve (red line, fit)



Towards differential polarization spectroscopy: the expected spectrum (blue line) is compared to the actual signal (red line, from the graph on the right)



Advantages of polarization spectroscopy

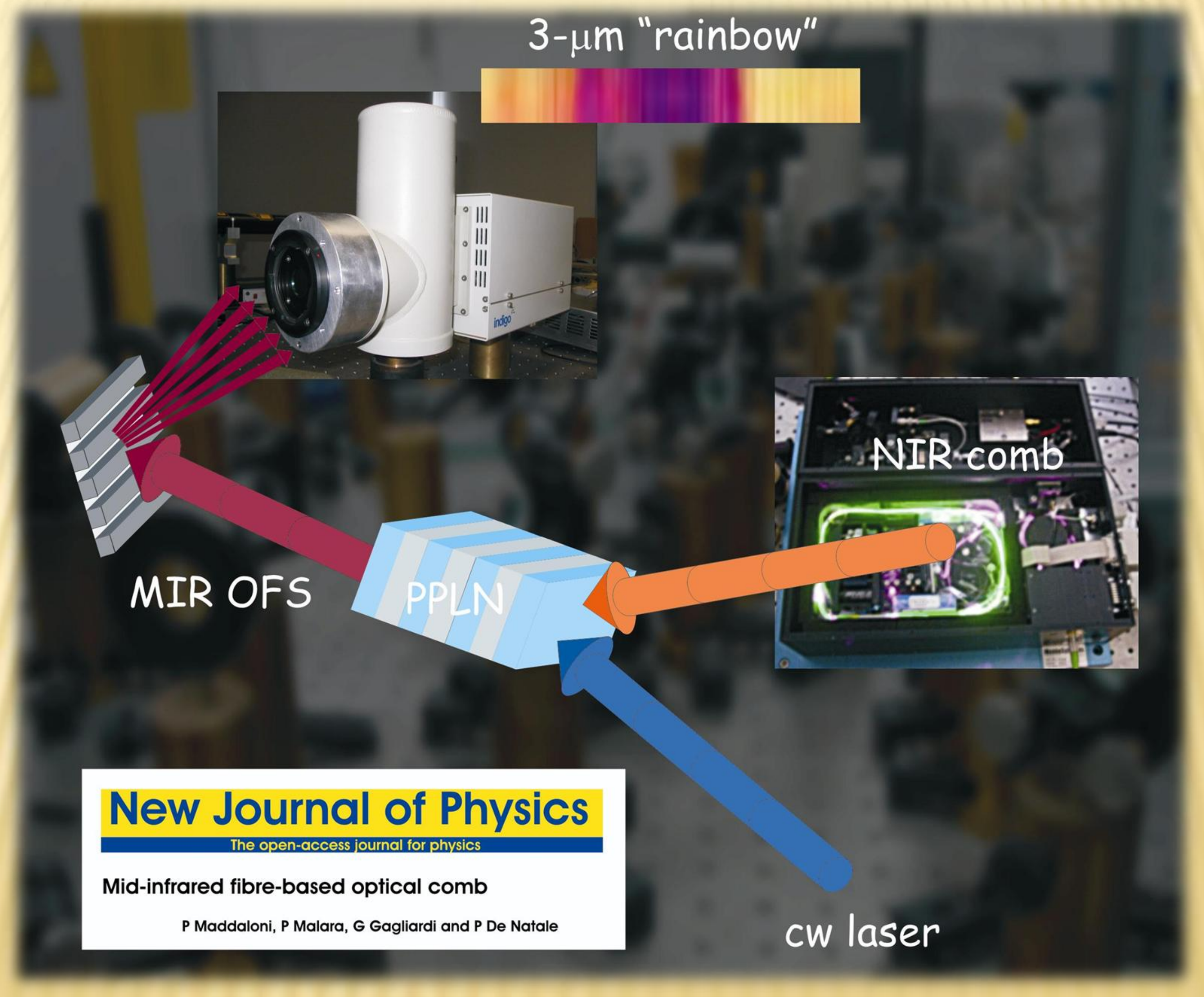
Polarization spectroscopy allows to obtain dispersion-like signals without any laser frequency modulation

- no need of high-frequency modulation of the laser current: safer use, no limits on the correction speed
- no need of signal demodulation: no limits in the locking bandwidth due to the lock-in amplifier time constant
- very sensitive detection
- possibility to obtain a zero background signal with a differential acquisition

These advantages have to be supported by:

- an improved mechanical isolation of the apparatus
- a better coupling of the correction signal to the laser

DOWN-CONVERTING A COMB TO MID-IR



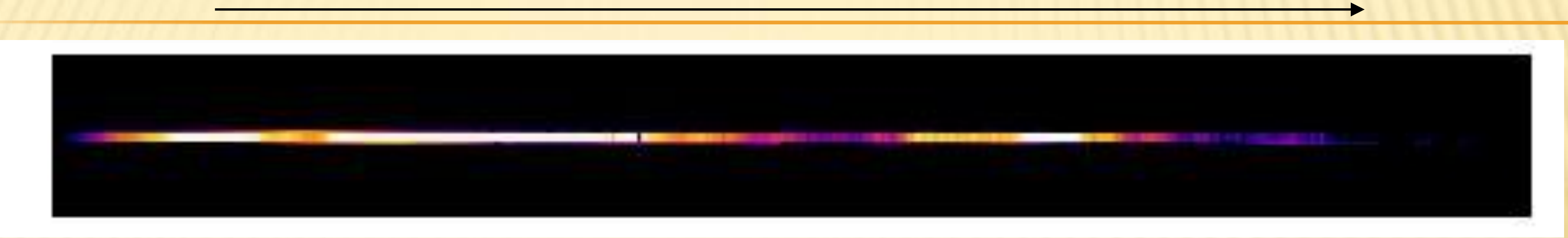
New Journal of Physics

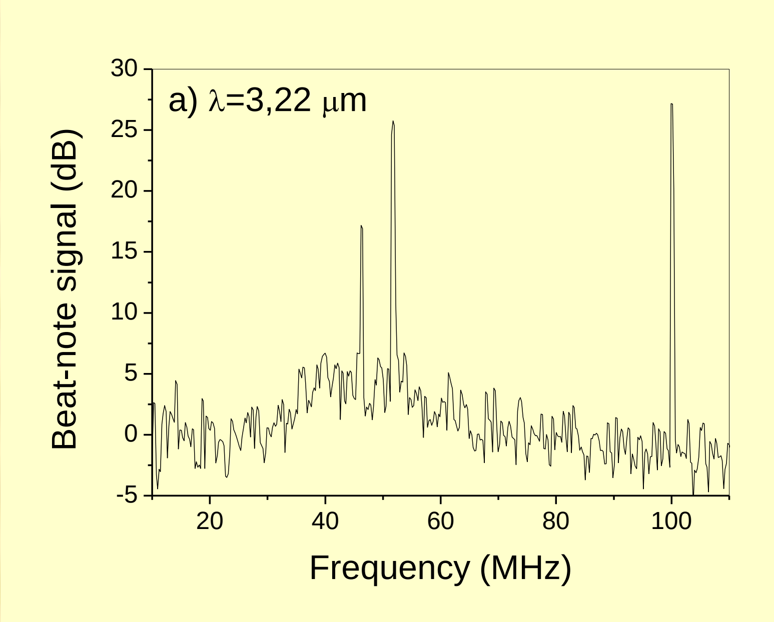
The open-access journal for physics

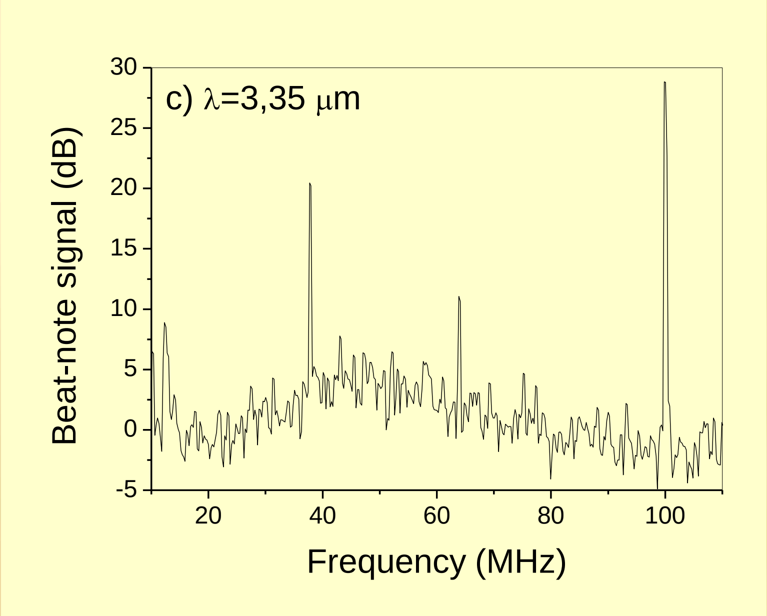
Mid-infrared fibre-based optical comb

P Maddaloni, P Malara, G Gagliardi and P De Natale

The 3 micron comb



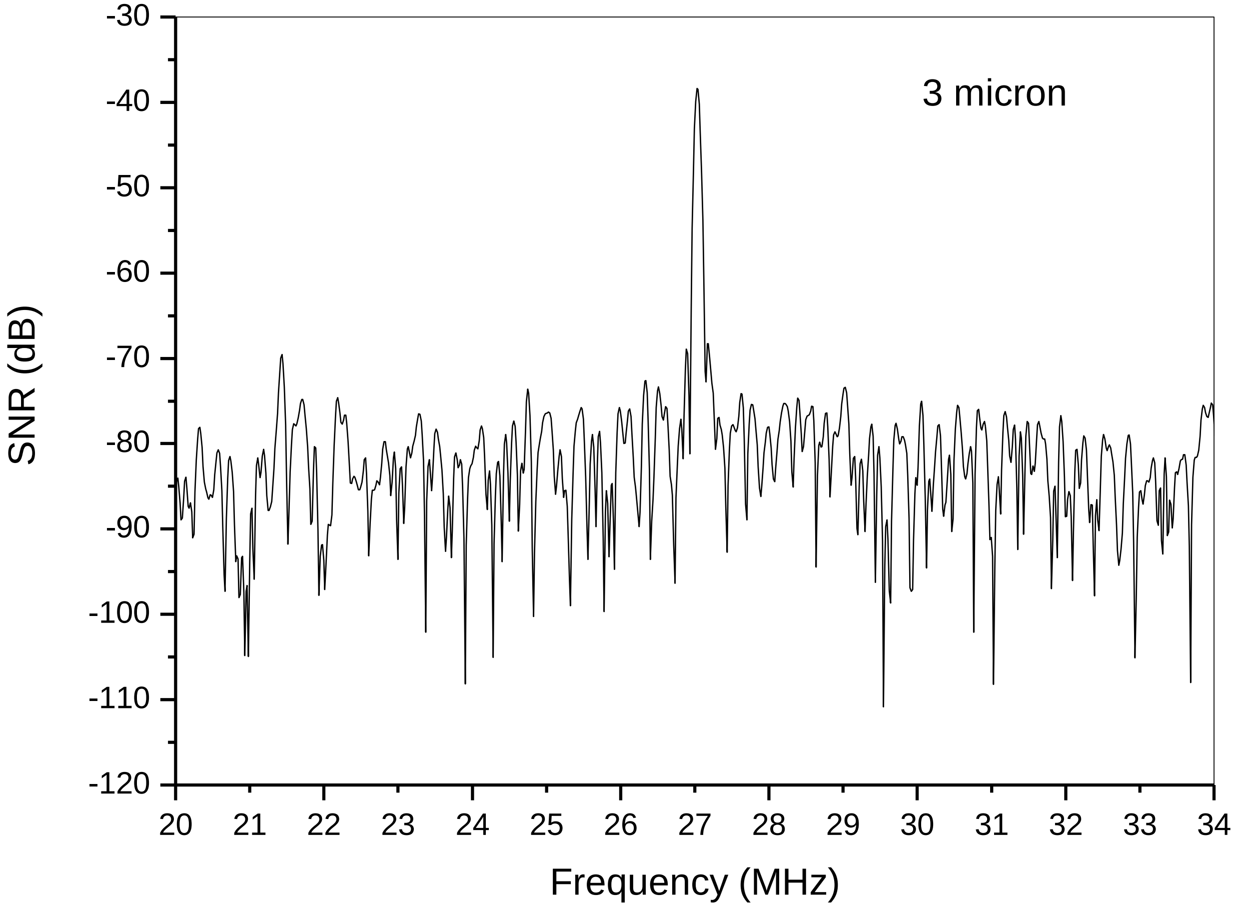




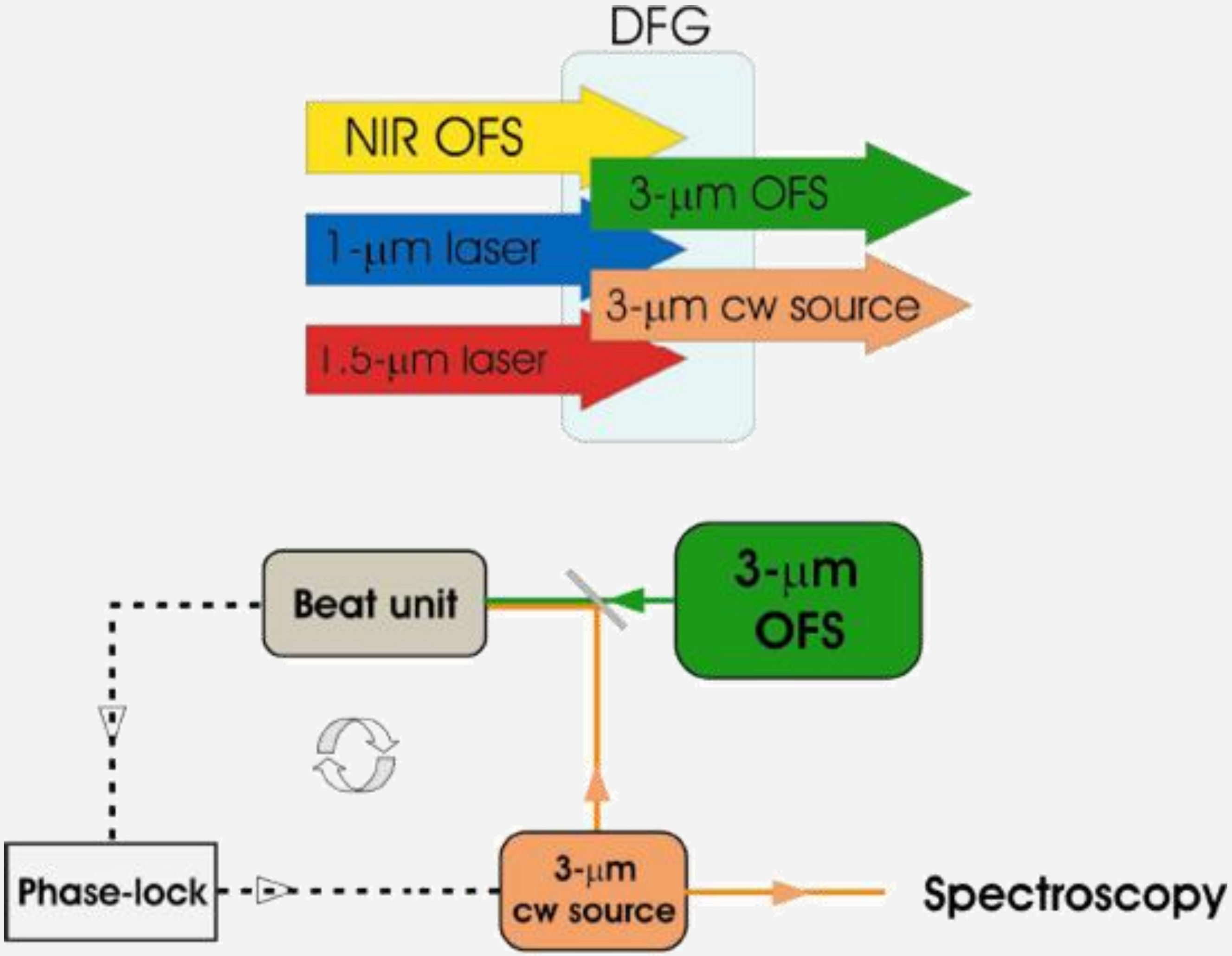
- P_{tot}=10 μW
- N=45000
- P_{tooth}=200 pW

Maddaloni et al., *New Journal of Physics* **8**, 262 (2006).

BEAT-NOTE DIRECTLY AT 3-μm WAVELENGTH



DIRECT LASER REFERENCING TO A MID-IR FREQUENCY COMB

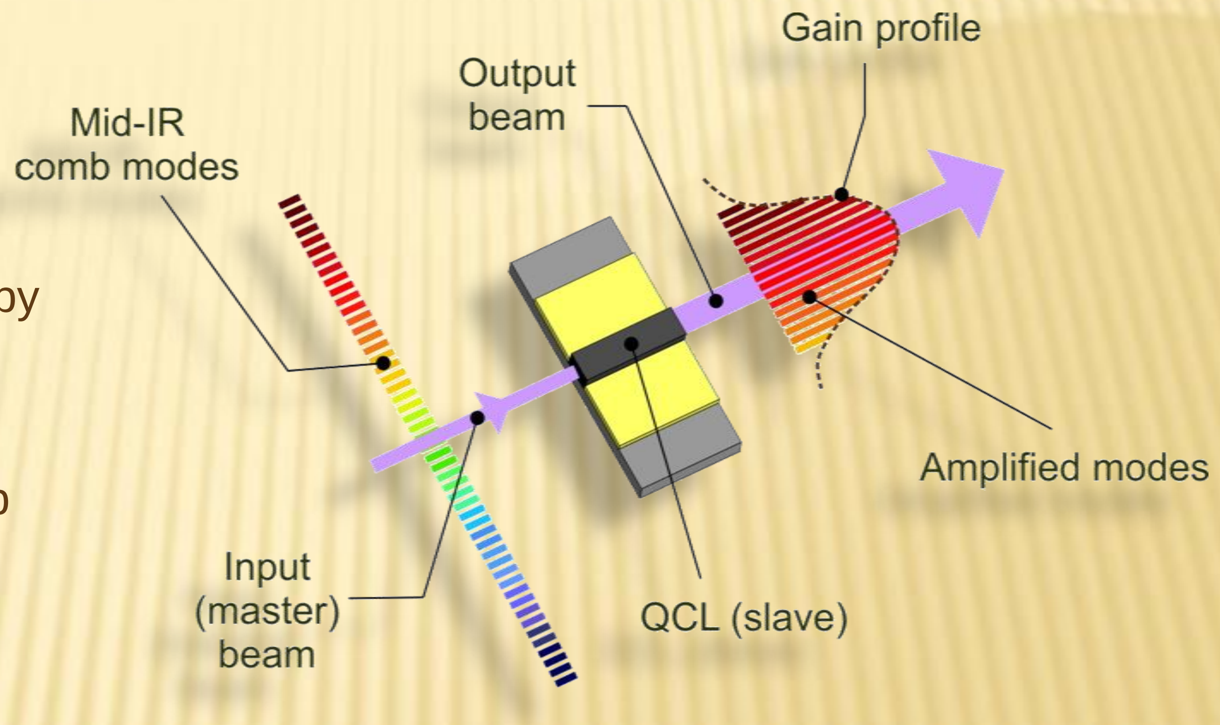


Malara et al., ***Opt. Exp.***, **16**, 8242-8249 (2008)

Comb amplification by QCLs

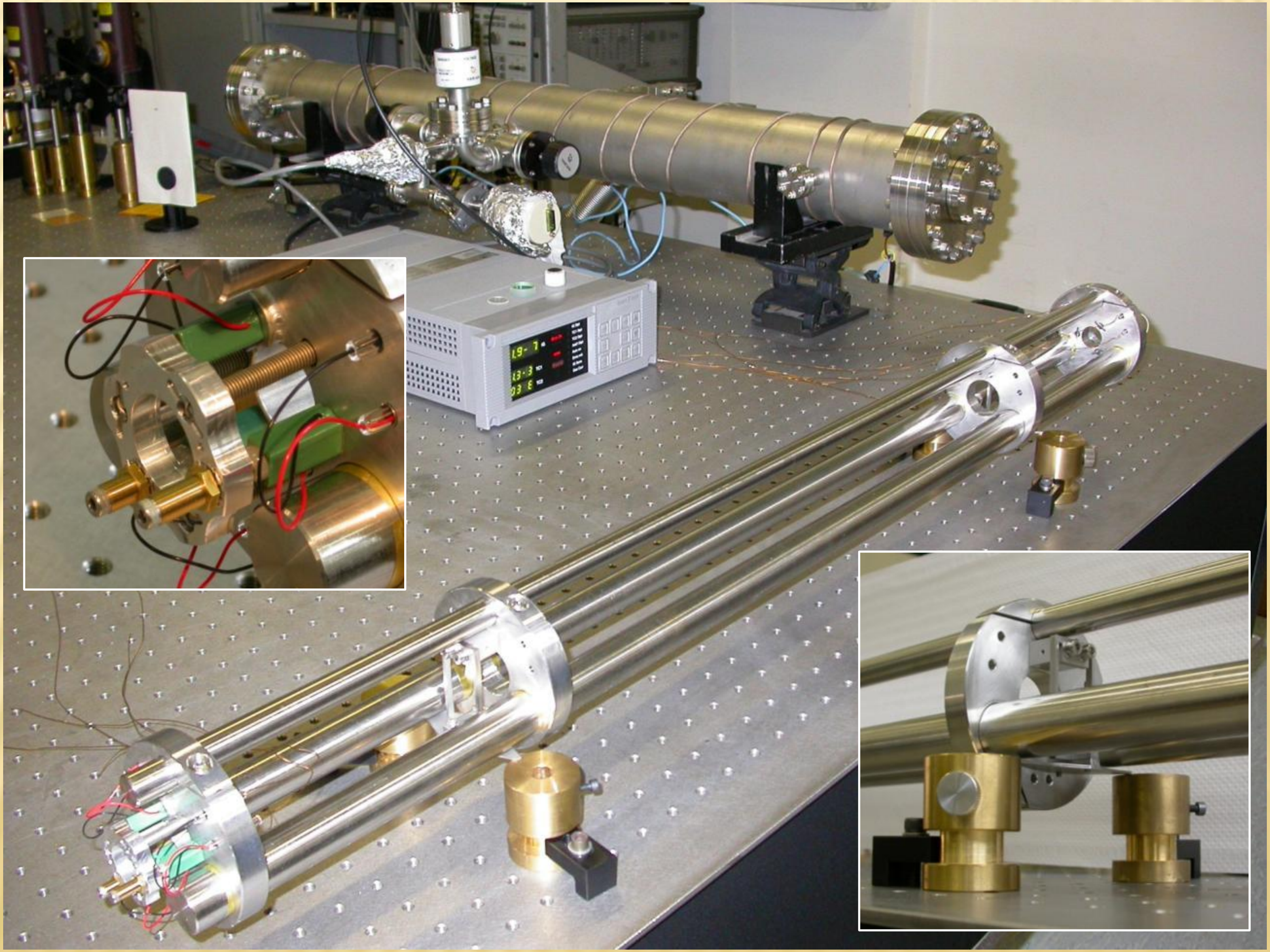
Some applications:

- Direct MIR Comb Spectroscopy
- Multispectral MIR imaging
- Cavity-enhanced Direct Comb Spectroscopy

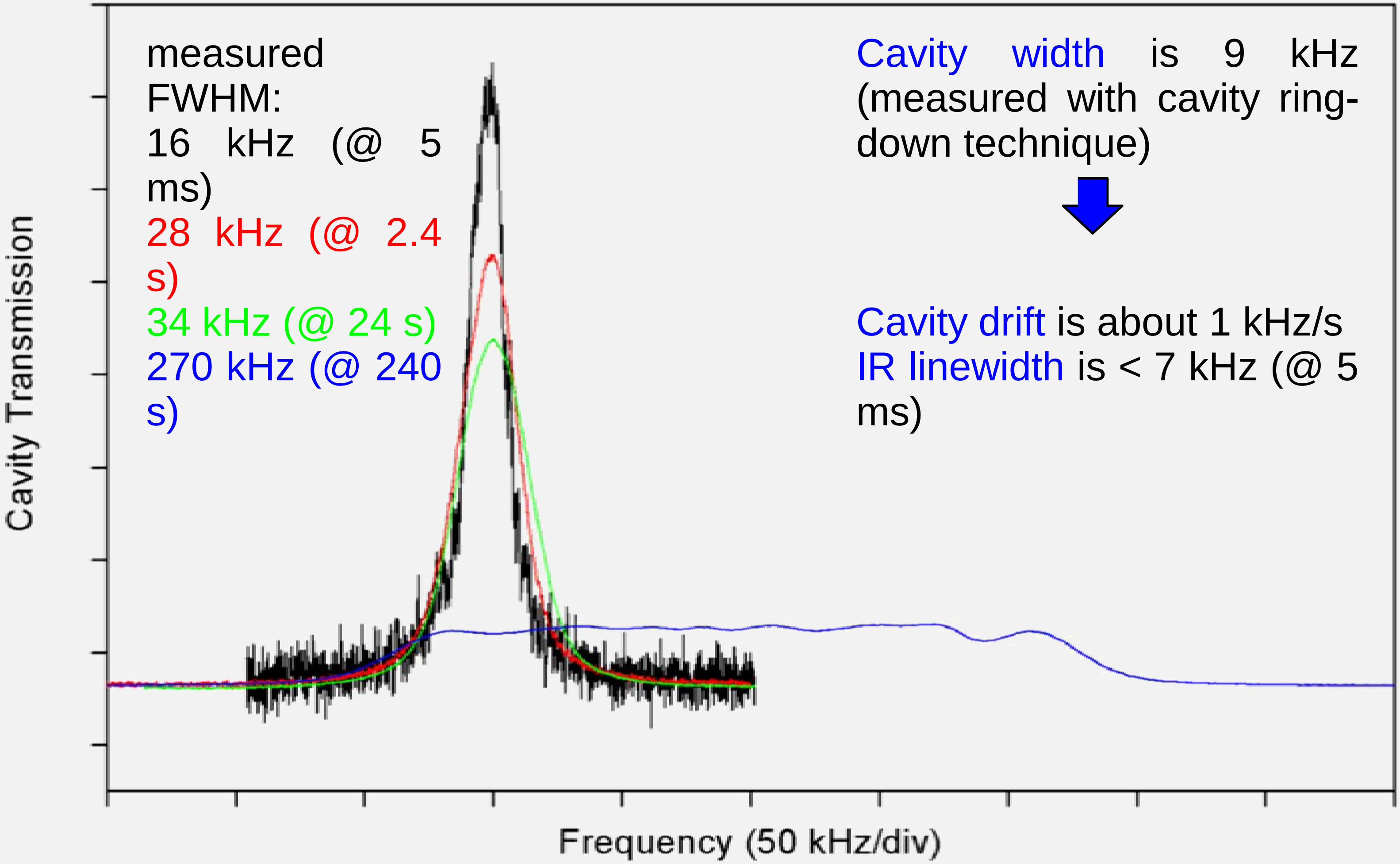


High-finesse Fabry-Perot cavity

F > 25000 @ 4.5 μm
FSR = 150 MHz



IR - cavity relative stability



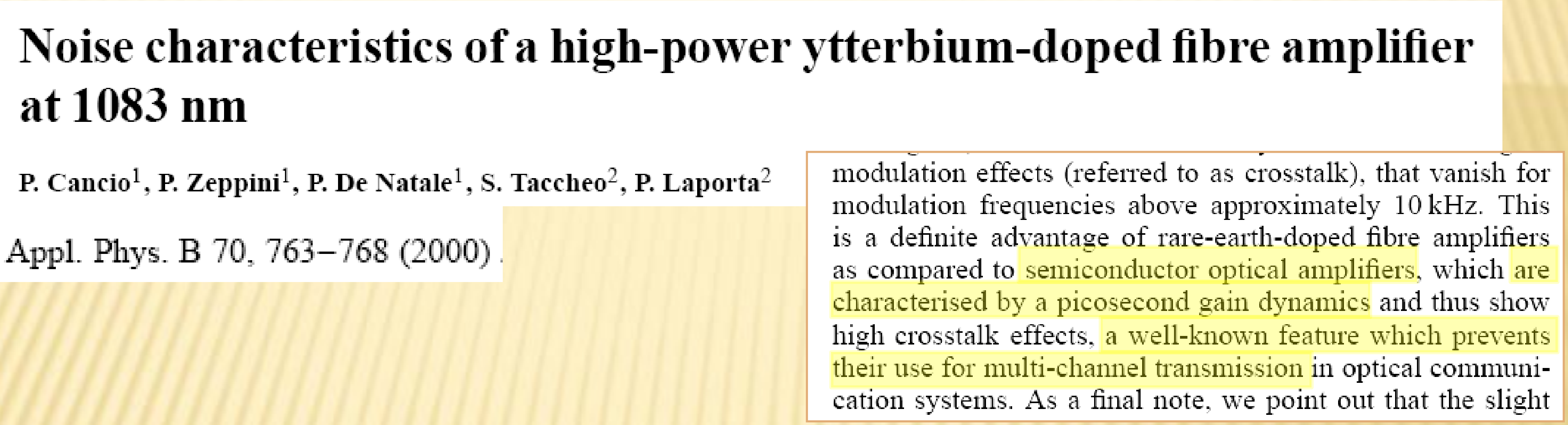
measured FWHM:
16 kHz (@ 5 ms)
28 kHz (@ 2.4 s)
34 kHz (@ 24 s)
270 kHz (@ 240 s)

Cavity width is 9 kHz (measured with cavity ring-down technique)

⬇

Cavity drift is about 1 kHz/s
IR linewidth is < 7 kHz (@ 5 ms)

Comb amplification

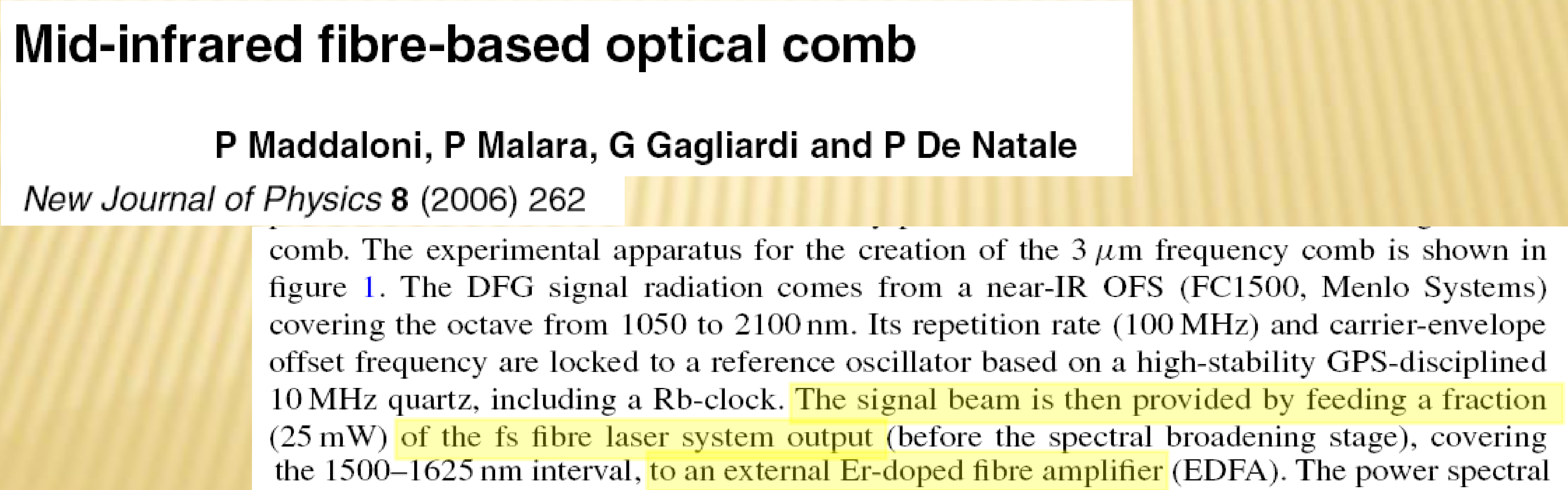


Noise characteristics of a high-power ytterbium-doped fibre amplifier at 1083 nm

P. Cancio[1], P. Zeppini[1], P. De Natale[1], S. Taccheo[2], P. Laporta[2]

Appl. Phys. B 70, 763–768 (2000)

modulation effects (referred to as crosstalk), that vanish for modulation frequencies above approximately 10 kHz. This is a definite advantage of rare-earth-doped fibre amplifiers as compared to semiconductor optical amplifiers, which are characterised by a picosecond gain dynamics and thus show high crosstalk effects, a well-known feature which prevents their use for multi-channel transmission in optical communication systems. As a final note, we point out that the slight



Mid-infrared fibre-based optical comb

P Maddaloni, P Malara, G Gagliardi and P De Natale

New Journal of Physics **8** (2006) 262

comb. The experimental apparatus for the creation of the 3 μm frequency comb is shown in figure 1. The DFG signal radiation comes from a near-IR OFS (FC1500, Menlo Systems) covering the octave from 1050 to 2100 nm. Its repetition rate (100 MHz) and carrier-envelope offset frequency are locked to a reference oscillator based on a high-stability GPS-disciplined 10 MHz quartz, including a Rb-clock. The signal beam is then provided by feeding a fraction (25 mW) of the fs fibre laser system output (before the spectral broadening stage), covering the 1500–1625 nm interval, to an external Er-doped fibre amplifier (EDFA). The power spectral

Comb amplification by semiconductors

Single mode amplification of near-IR combs by injection locking into semiconductor devices has been already performed in several experiments. See for example:

- *H. S. Moon et al.*, Appl. Phys. Lett. **89**, 181110 (2006)

Selection and amplification of the components of an optical frequency comb using a femtosecond laser injection-locking technique in a near-IR Fabry-Perot diode laser

- *S. E. Park et al.*, Opt. Lett. **31**, 3594 (2006)

A distributed-Bragg-reflector near-IR semiconductor laser is injection locked by a single component of an optical frequency comb

- *F. C. Cruz et al.*, Opt. Lett. **31**, 1337 (2006)

A tapered semiconductor amplifier is injection seeded by a femtosecond optical frequency comb at 780 nm from a mode-locked Ti:sapphire laser. A spectral window of super-continuum light generated in a photonic fiber has also been amplified.

- *M. Laemmlin et al.*, Electr. Lett. **42** (2006)

Distortion-free optical amplification of 20-80 GHz modelocked laser pulses at 1.3 μ*m using quantum dots*

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