

Cavity enhanced optical frequency comb spectroscopy

Lucile Rutkowski

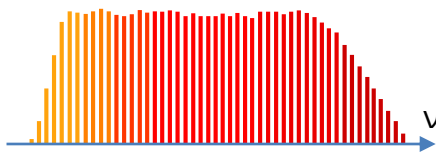
Univ Rennes, CNRS, IPR (Institut de Physique de Rennes)-UMR 6251, F-35000 Rennes, France

lucile.rutkowski@univ-rennes1.fr

Introduction

High resolution AND broad bandwidth
 Efficient coupling with an **enhancement cavity**

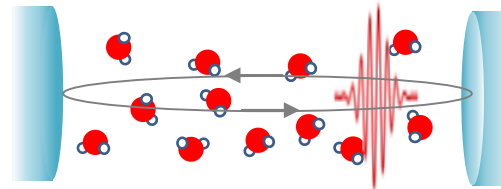
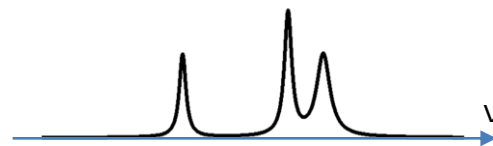
Comb spectrum



Mode locked laser

~10⁵ equidistant cw lasers

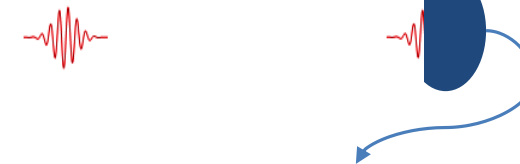
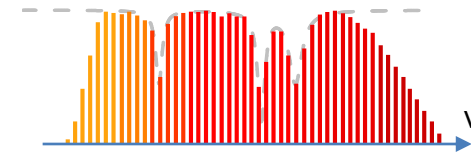
Molecular absorption



Enhancement cavity
Sensitivity

Interaction length
 \propto **finesse**

Spectral transmission



Broadband detection

Vernier
FTS
VIPA
DCS

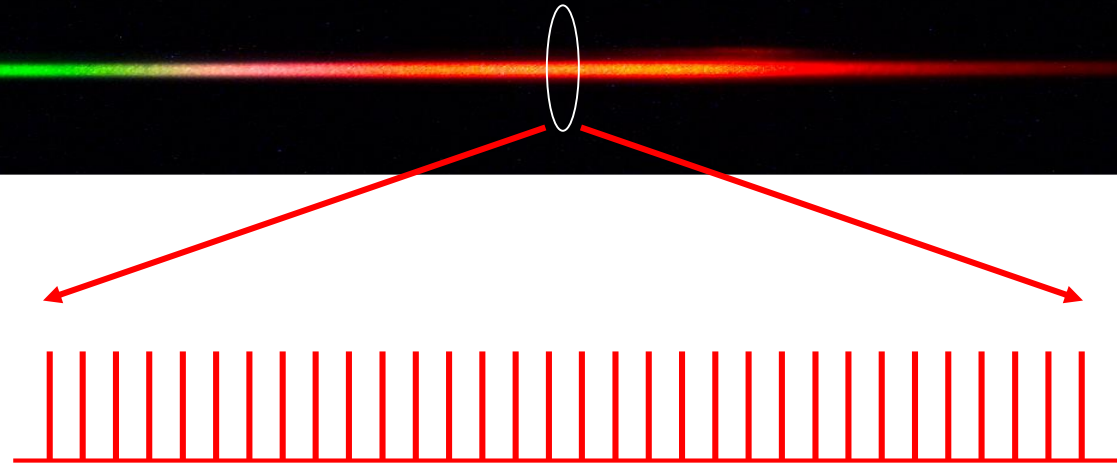
...

Measure entire absorption bands and/or different species **simultaneously**, with **high sensitivity** and in **short acquisition times**



Optical frequency combs

The bandwidth of incoherent light sources with the resolution of cw lasers



Thousands of synchronized laser lines

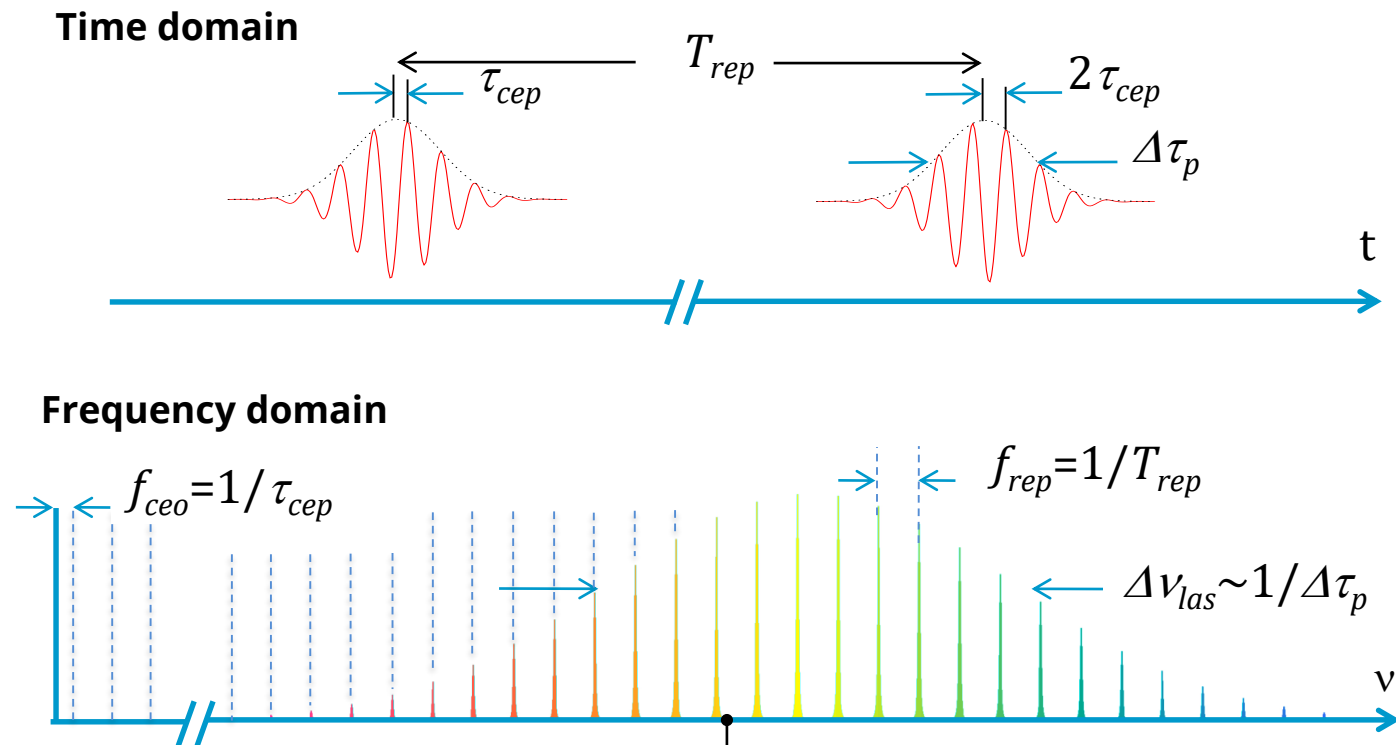


Prix Nobel 2005

John Hall & Theodor Hänsch
for their contributions to the development
of laser-based precision spectroscopy,
including the optical frequency comb technique

From pulsed lasers to combs

Mode locked femtosecond lasers



n	Mode index	$10^4 - 10^6$
f_{rep}	Repetition rate	50 MHz – 10 GHz
f_{ceo}	Carrier-envelop offset frequency	$0 - f_{rep}$

Optical frequency of the n^{th} mode:

$$\nu_n = n f_{rep} + f_{ceo}$$

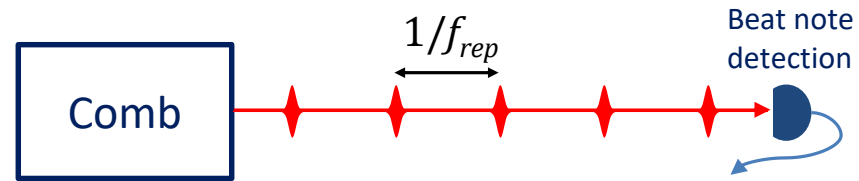
The perfect link between radio and optical frequency ranges

Stabilization

Repetition rate

Measurement: *fast detector*

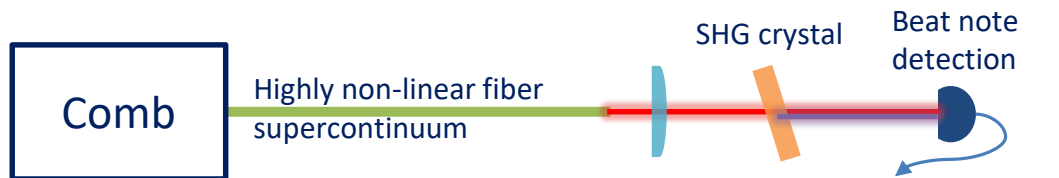
Actuators: *cavity length (stepper motor, PZT, EOM) and/or pump current*



Offset frequency

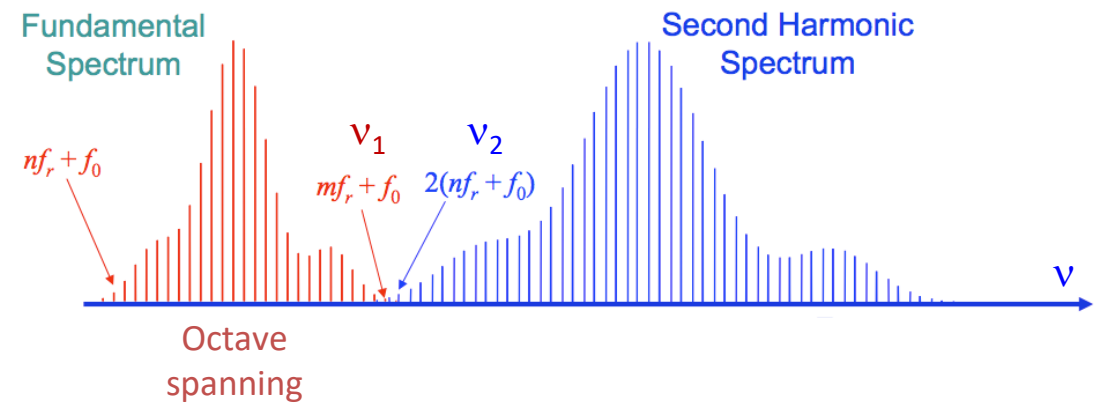
Measurement: *f-2f interferometry*

Actuators: *oscillator pump current, intracavity dispersion and/or external A/EOM*



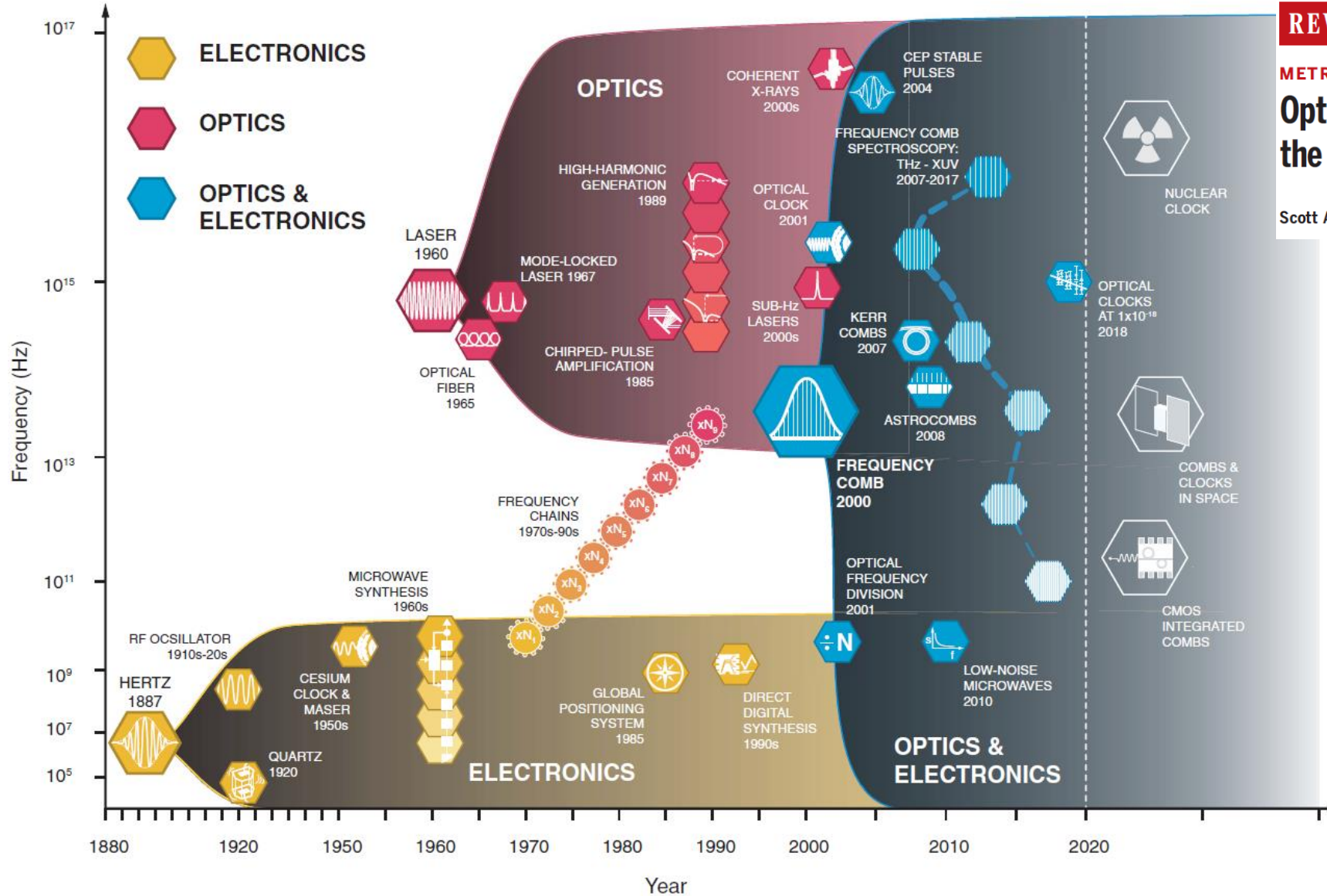
$$m = 2n$$

$$\nu_2 - \nu_1 = 2(nf_{\text{rep}} + f_0) - (mf_{\text{rep}} + f_0) = f_0$$





Opto-electronics



REVIEW SUMMARY

METROLOGY

Optical frequency combs: Coherently uniting the electromagnetic spectrum

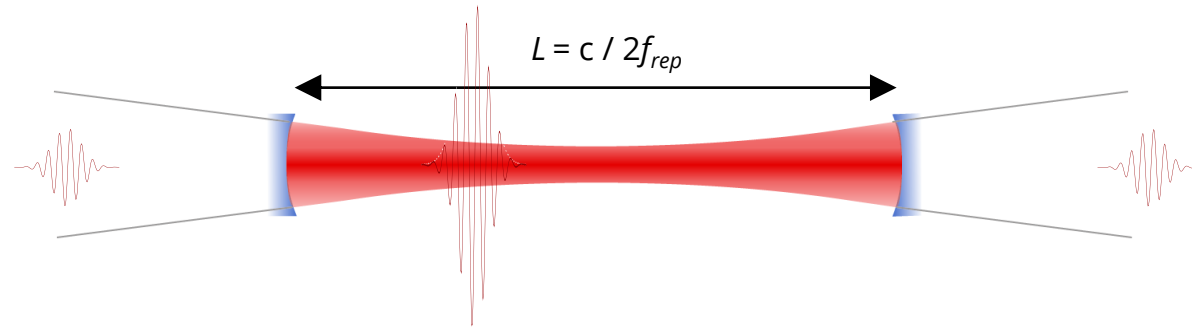
Scott A. Diddams*, Kerry Vahala*, Thomas Udem*

Science, 369, 267 (2020)

Cavity enhancement

Perfect comb-cavity matching

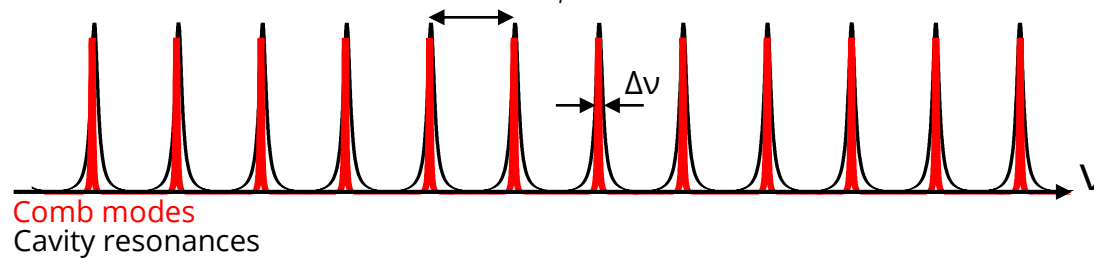
Time domain Match the cavity length to the pulse separation



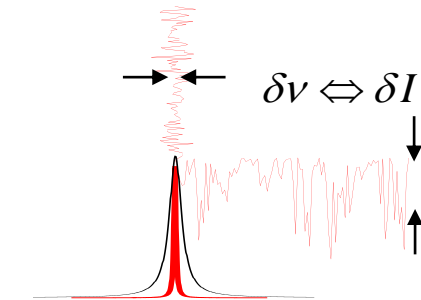
Frequency domain Match the cavity resonances to the comb modes

Cavity finesse: $F = FSR / \Delta\nu$

Cavity free spectral range: $FSR = c/2nL = f_{rep}$

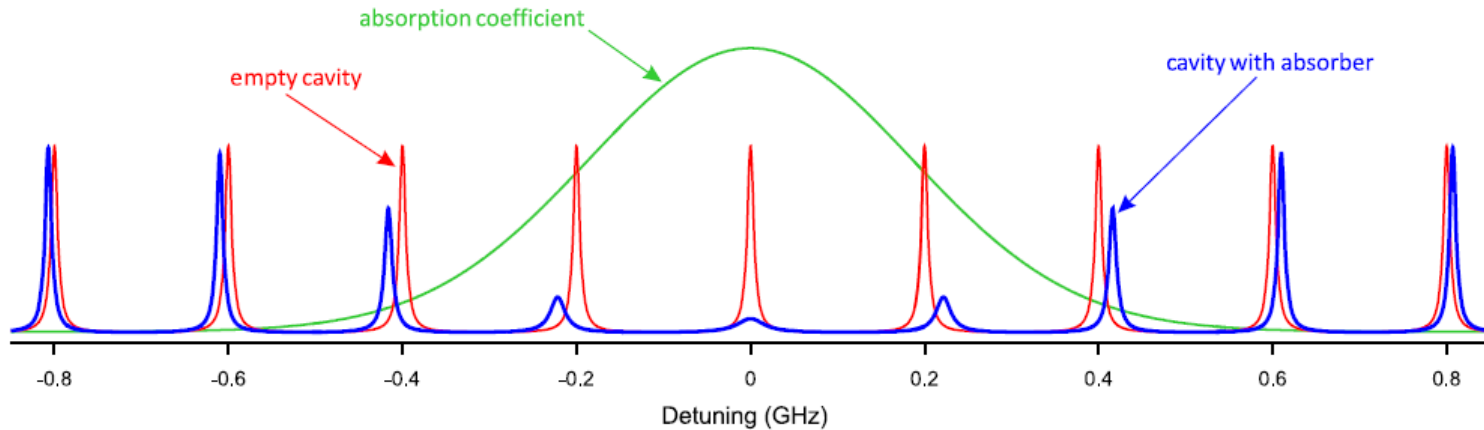


Frequency to amplitude noise conversion



Intra-cavity absorption

Molecular transition = absorption + local variation of the refractive index



Amplitude reduction

Resonance broadening

Shift of the resonant frequencies

One-dimensional frequency-based spectroscopy

Opt. Express 23(11), 14472 (2015)

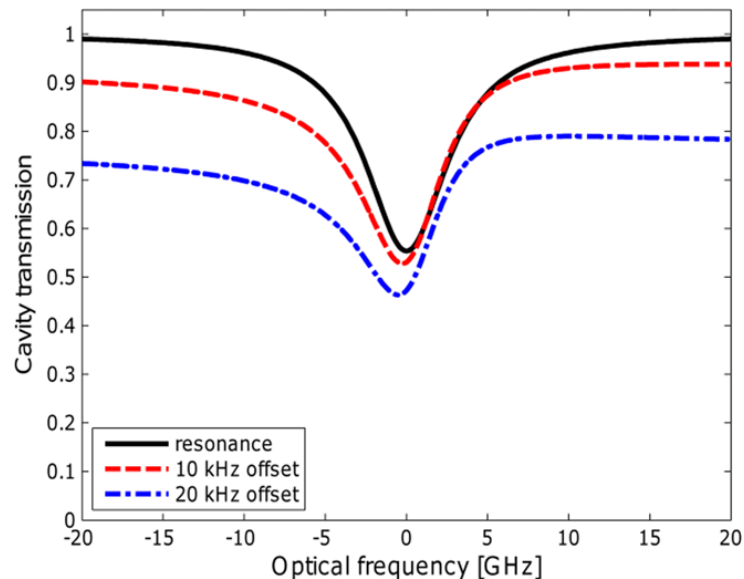
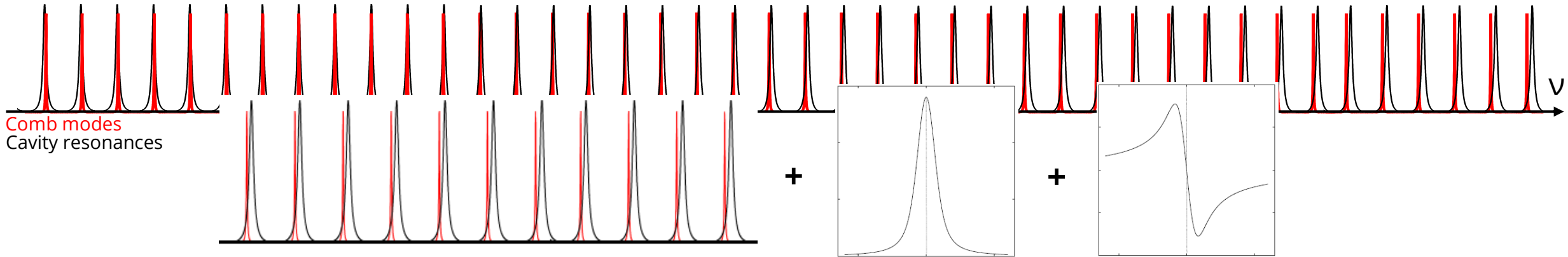
Agata Cygan,^{1,*} Piotr Wcislo,¹ Szymon Wójtewicz,¹ Piotr Masłowski,¹
Joseph T. Hodges,² Roman Ciuryło,¹ and Daniel Lisak¹

¹Institute of Physics, Faculty of Physics, Astronomy and Informatics, Nicolaus Copernicus University in Torun, Grudziadzka 5, 87-100 Torun, Poland.

²National Institute of Standards and Technology, 100 Bureau Drive, Gaithersburg, Maryland 20899, USA

Intra-cavity dispersion

The FSR- f_{rep} matching cannot be perfect on the entire comb bandwidth.



Appl Phys B (2013) 110:163–175
DOI 10.1007/s00340-012-5024-7

Applied Physics B
Lasers and Optics

Cavity-enhanced optical frequency comb spectroscopy in the mid-infrared application to trace detection of hydrogen peroxide

A. Foltynowicz · P. Masłowski · A.J. Fleisher · B.J. Bjork · J. Ye



Contents lists available at ScienceDirect

Journal of Molecular Spectroscopy

journal homepage: www.elsevier.com/locate/jms



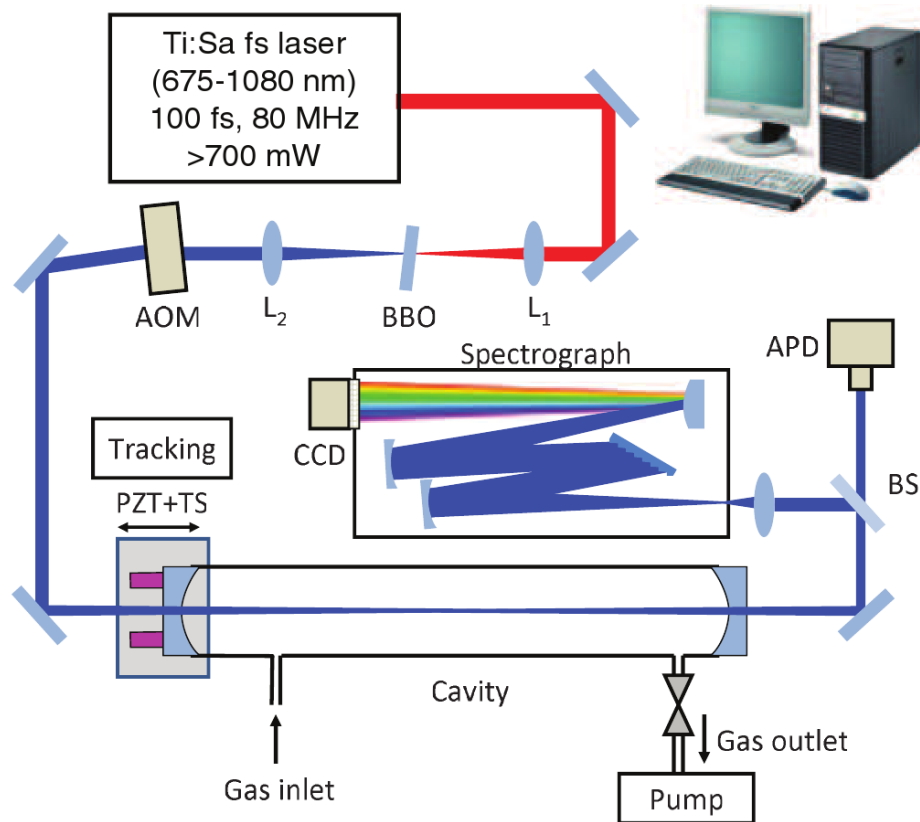
Quantitative modeling of complex molecular response in coherent cavity-enhanced dual-comb spectroscopy

Adam J. Fleisher*, David A. Long, Joseph T. Hodges

Material Measurement Laboratory, National Institute of Standards and Technology, 100 Bureau Drive, Gaithersburg, MD 20899, USA



1D Dispersive spectrometers



1D dispersion

- Diffraction grating + CCD array
- Resolution > GHz
- Frequency calibration of the grating
- Spectral coverage – few nm
- Acquisition time – ms

Mode-locked cavity-enhanced absorption spectroscopy

Opt. Express 10(19), 1033 (2002)

Titus Gherman and Daniele Romanini

PHYSICAL REVIEW A 85, 051804(R) (2012)

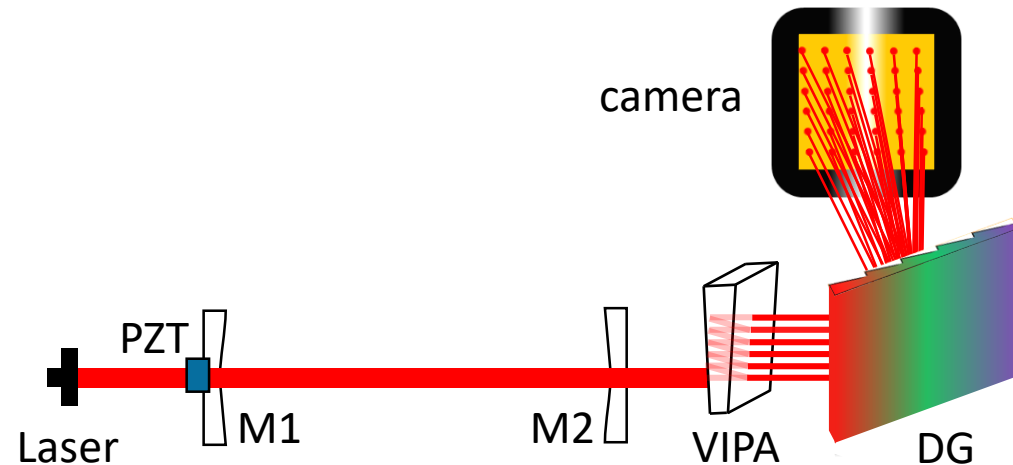
Cavity-enhanced multiplexed comb spectroscopy down to the photon shot noise

R. Grilli, G. Méjean, C. Abd Alrahman, I. Ventrillard, S. Kassi, and D. Romanini

Université de Grenoble 1 / CNRS, LIPhy UMR 5588, F-38041 Grenoble, France

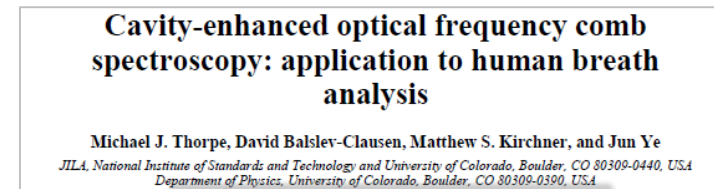
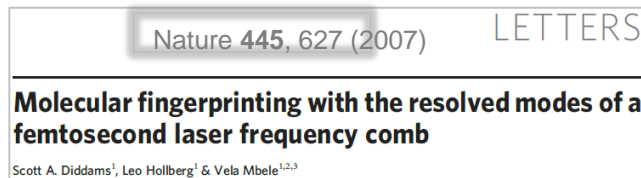
(Received 8 February 2012; published 22 May 2012)

2D Dispersive spectrometers

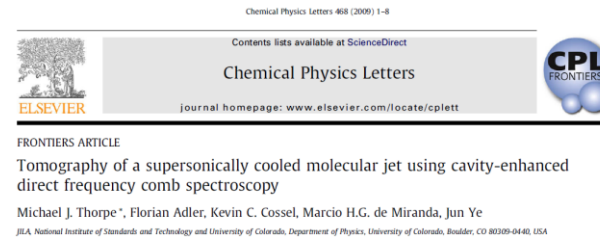


VIPA (NIR and MIR)

- Virtually Imaged Phased Array + orthogonal dispersion grating + camera
- Mode per mode resolution possible
- # spectral elements ~ # of pixels
- Acquisition time – ms



Opt. Express 16, 2387 (2008)



Mid-Infrared Time-Resolved Frequency Comb Spectroscopy of Transient Free Radicals

Adam J. Fleisher,^{1,2} Bryce J. Bjork,¹ Thinh Q. Bui,³ Kevin C. Cossel,¹ Mátthio Okumura,^{4,5} and Jun Ye^{1,2}

¹JILA, National Institute of Standards and Technology and University of Colorado, Department of Physics, 440 UCB, Boulder, Colorado 80309, United States

²Material Measurement Laboratory, National Institute of Standards and Technology, 100 Bureau Drive, Gaithersburg, Maryland 20899, United States

³Arthur Amos Noyes Laboratory of Chemical Physics, Division of Chemistry and Chemical Engineering, California Institute of Technology, 1200 East California Boulevard, Pasadena, California 91125, United States

SCIENTIFIC REPORTS

OPEN Broadband Optical Cavity Mode Measurements at Hz-Level Precision With a Comb-Based VIPA Spectrometer

Grzegorz Kowzan, Dominik Charczun, Agata Cygan, Ryszard S. Trawiński, Daniel Lisak & Piotr Masłowski

Received: 18 December 2018
Accepted: 8 May 2019
Published online: 03 June 2019

MOLECULAR PHYSICS
2020, VOL. 118, NO. 16, e1733116 (9 pages)
<https://doi.org/10.1080/00268976.2020.1733116>

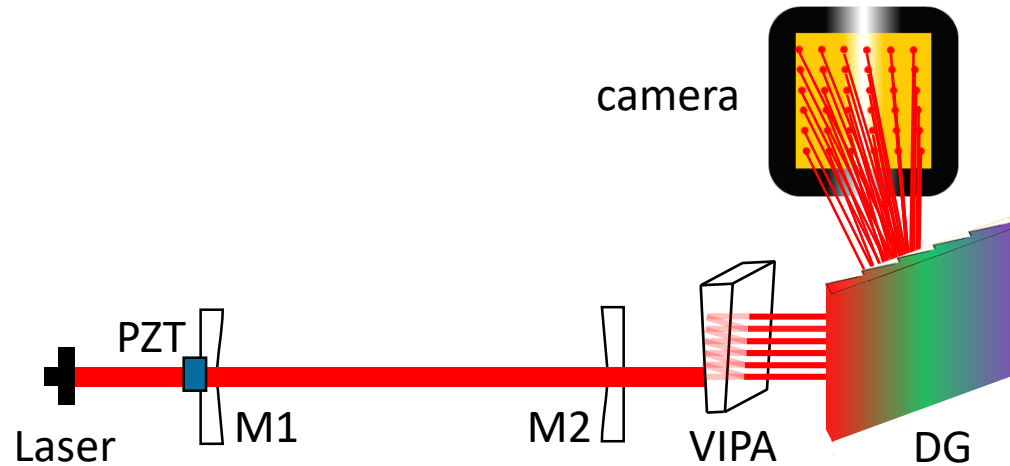
NEW VIEW

A rapid, spatially dispersive frequency comb spectrograph aimed at gas phase chemical reaction kinetics

Frances C. Roberts, H. J. Lewandowski, Billy F. Hobson and Julia H. Lehman

¹School of Chemistry, University of Leeds, Leeds, UK; ²JILA and Department of Physics, University of Colorado and the National Institute of Standards and Technology, Boulder, CO, USA

2D Dispersive spectrometers



VIPA (NIR and MIR)

- Virtually Imaged Phased Array + orthogonal dispersion grating + camera
- Mode per mode resolution possible
- # spectral elements \sim # of pixels
- Acquisition time – ms

Immersion grating (10 μm)

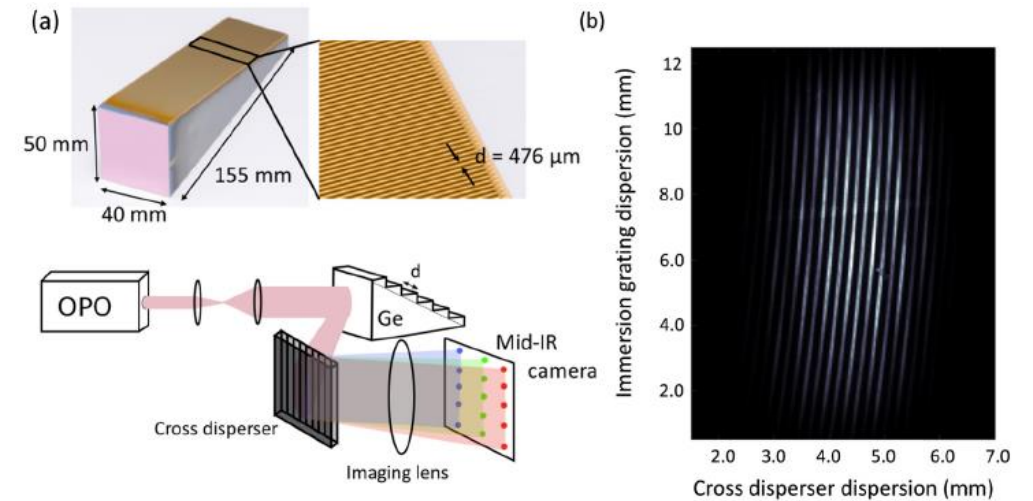


Comb-resolved spectroscopy with immersion grating in long-wave infrared

K. IWAKUNI,^{1,*} T. Q. BUI,¹ J. F. NIEDERMEYER,¹ T. SUKEGAWA,² AND J. YE¹

¹JILA, National Institute of Standards and Technology and University of Colorado, and Department of Physics, University of Colorado, Boulder, CO 80309, USA

²Canon Inc., 20-2, Kiyohara-Kogyodanchi, Utsunomiya, Tochigi 321-3292, Japan





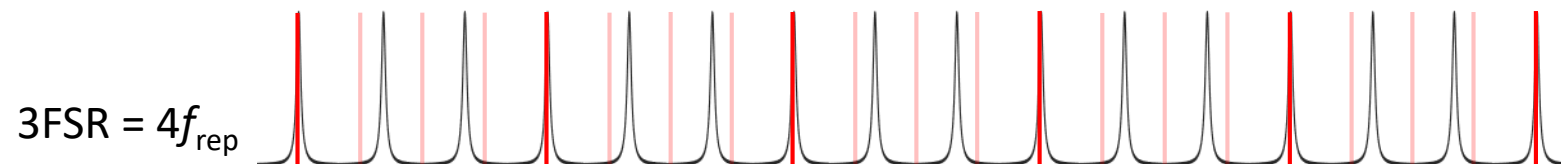
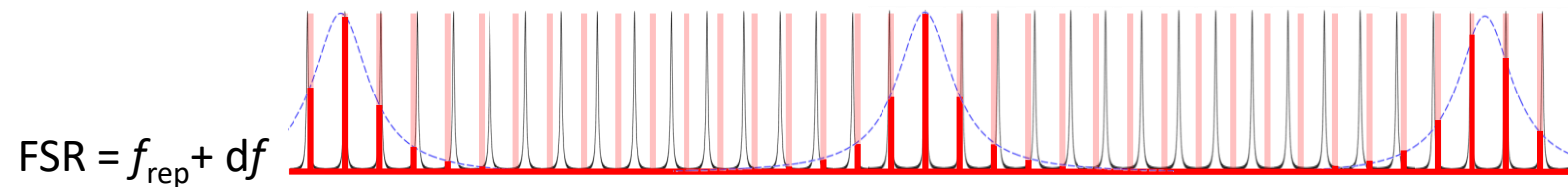
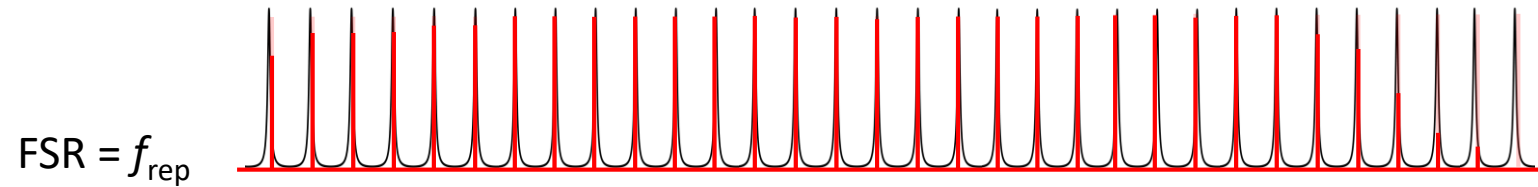
Vernier filtering approaches

- Use the cavity as a frequency filter
- Mismatch between FSR and f_{rep}

Review

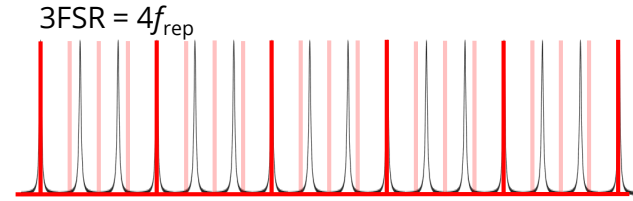
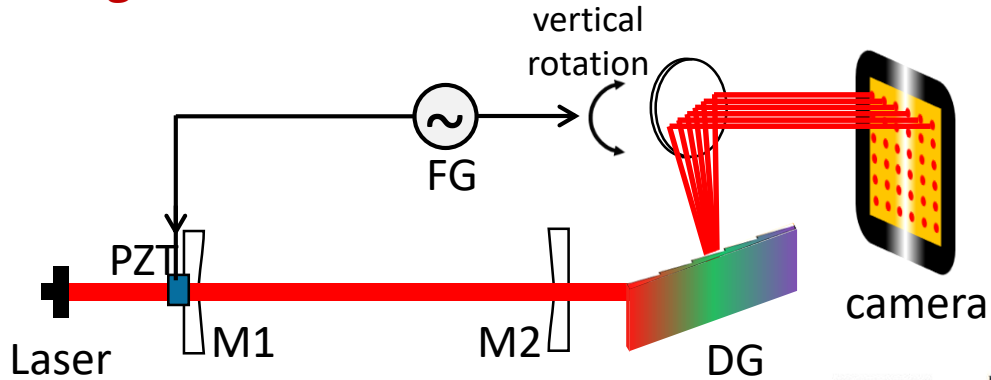
Cavity-Enhanced Frequency Comb Vernier Spectroscopy

Chuang Lu ¹, Jerome Morville ², Lucile Rutkowski ³ , Francisco Senna Vieira ¹ and Aleksandra Foltynowicz ^{1,*} 



Mode resolved Vernier

Using a femtosecond laser



Vernier in mode per mode filtering

- Spectral coverage : entire comb
- Comb mode resolution
- Acquisition time < sec
- Sensitivity limited by intensity noise

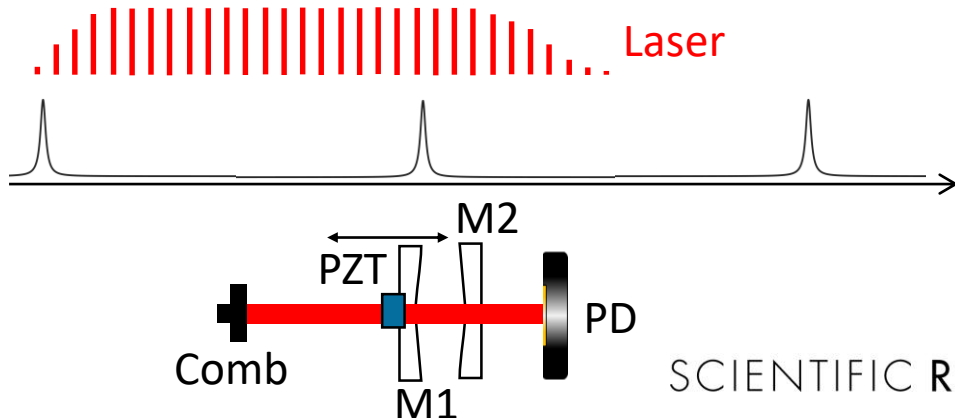
PHYSICAL REVIEW LETTERS
 PRL 99, 263902 (2007)
 Frequency Comb Vernier Spectroscopy for Broadband, High-Resolution, High-Sensitivity Absorption and Dispersion Spectra
 Christoph Gebke,¹ Björn Stein,¹ Albert Schliesser, Thomas Udem, and Theodor W. Hänsch
 Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Strasse 1, D-85748 Garching, Germany



Cavity-Enhanced Vernier Spectroscopy with a Chip-Scale Mid-Infrared Frequency Comb

Lukasz A. Sterczewski,^{1,2} Tzu-Ling Chen,¹ Douglas C. Ober, Charles R. Markus, Chadwick L. Canedy, Igor Vurgafman, Clifford Frez, Jerry R. Meyer, Mitchio Okumura,³ and Mahmood Bagheri⁴

Using micro-cavities

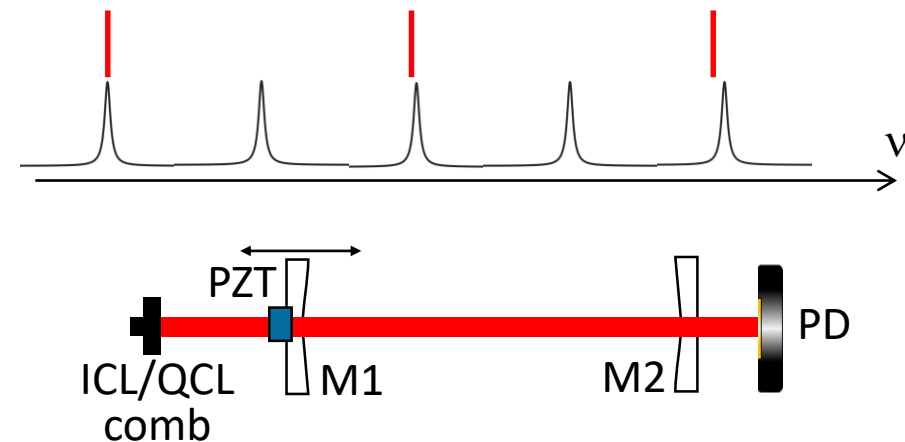


SCIENTIFIC REPORTS

Scanning micro-resonator direct-comb absolute spectroscopy

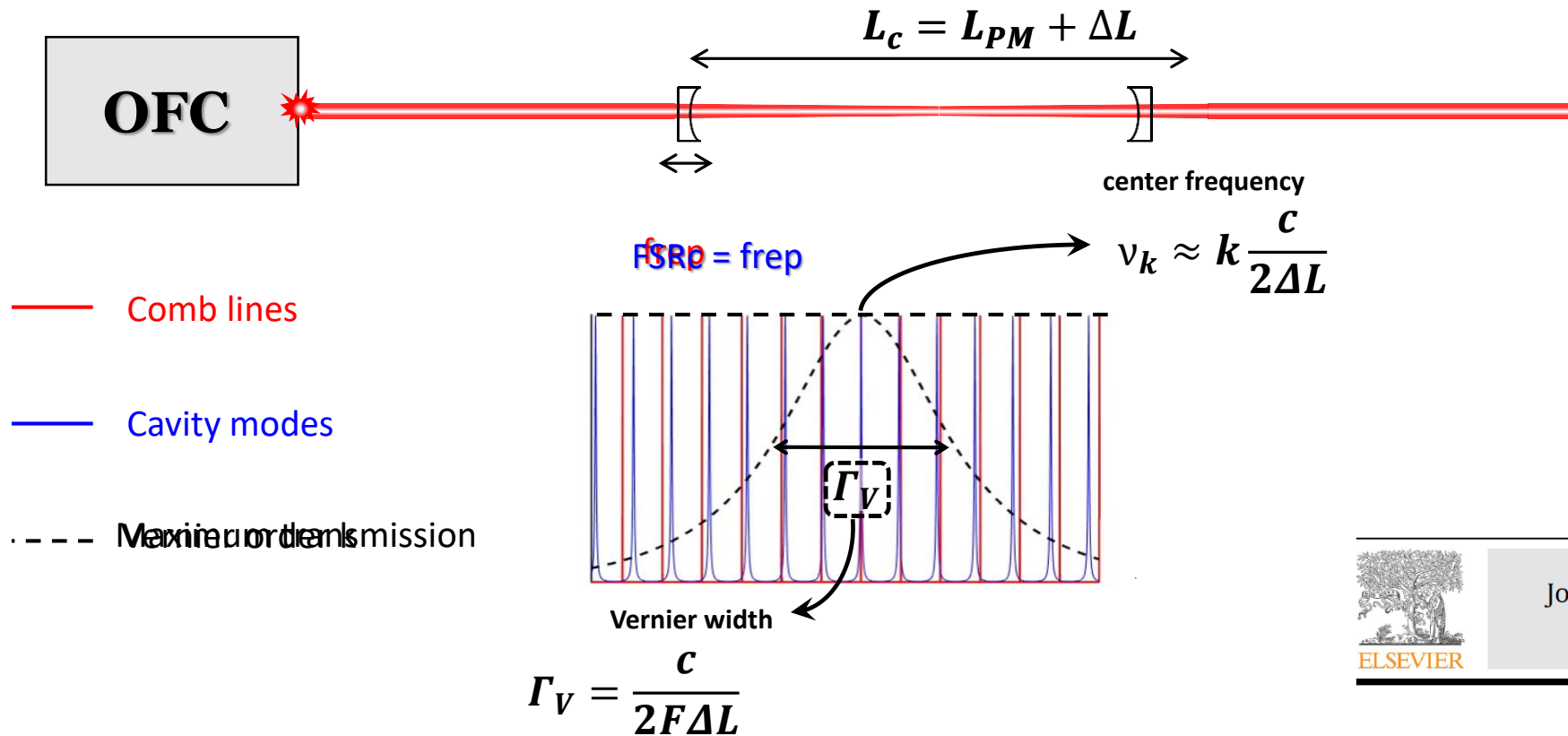
Alessio Gambetta^{1,2}, Marco Cassinero^{1,2}, Davide Gatti¹, Paolo Laporta^{1,2} & Gianluca Galzerano^{1,2}

Using QCL-combs





Continuous filtering



Journal of Quantitative Spectroscopy & Radiative Transfer 187 (2017) 204–214



Contents lists available at ScienceDirect
Journal of Quantitative Spectroscopy & Radiative Transfer
 journal homepage: www.elsevier.com/locate/jqsrt



Continuous Vernier filtering of an optical frequency comb for broadband cavity-enhanced molecular spectroscopy

Lucile Rutkowski^a, Jérôme Morville^b

^a Department of Physics, Umeå University, 901 87, Umeå, Sweden

^b Institut Lumière Matière, CNRS UMR5306, Université Lyon 1, Université de Lyon, 69622 Villeurbanne CEDEX, France

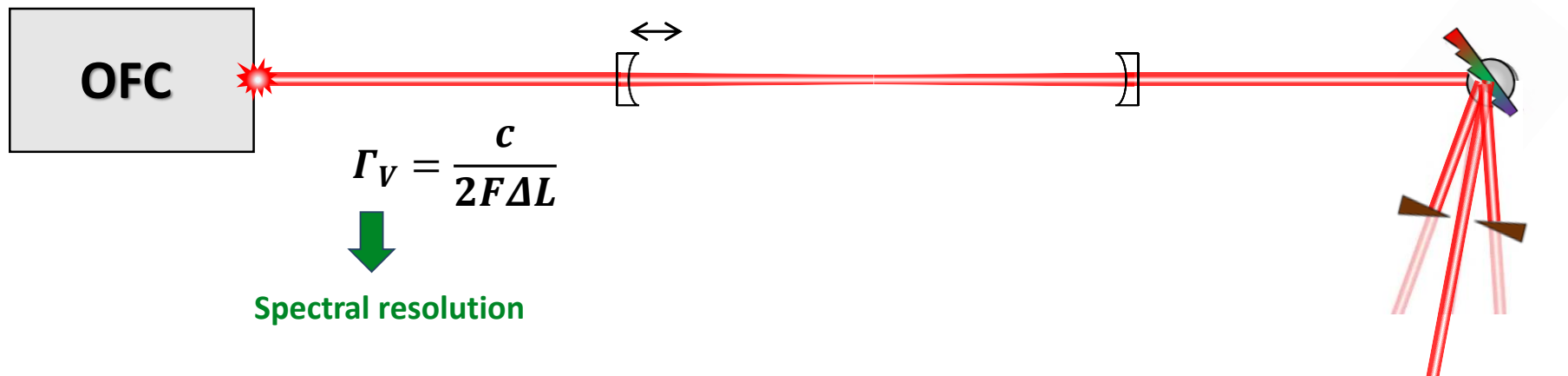
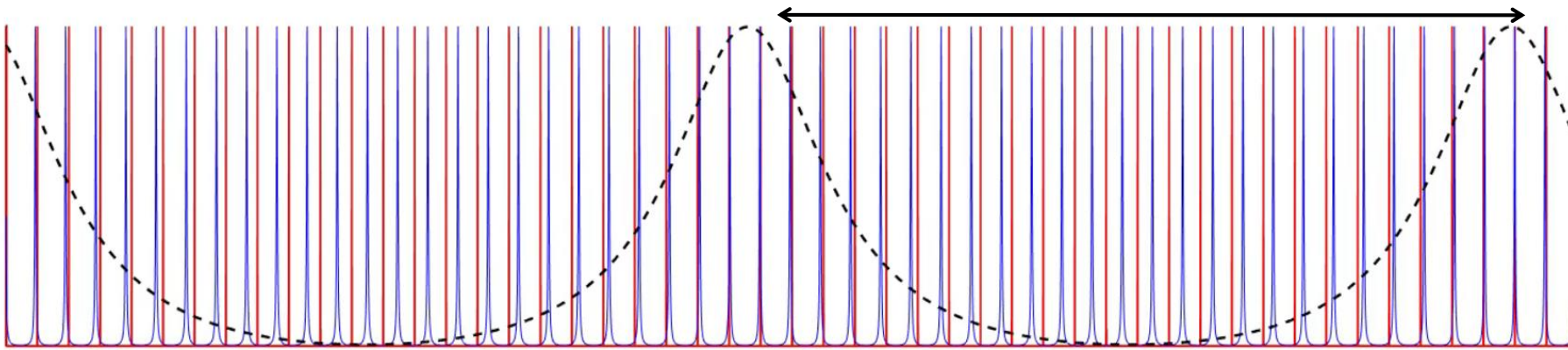


Continuous filtering

Vernier in continuous filtering

- Spectral coverage : entire comb
- Tunable resolution $> 5f_{rep}$
- Acquisition time \sim sec
- Immune to intensity noise
- Require external frequency calibration

$$FSR_V = \frac{L_{PM}}{\Delta L} FSR_c \rightarrow \text{THz}$$



Discharge spectroscopy

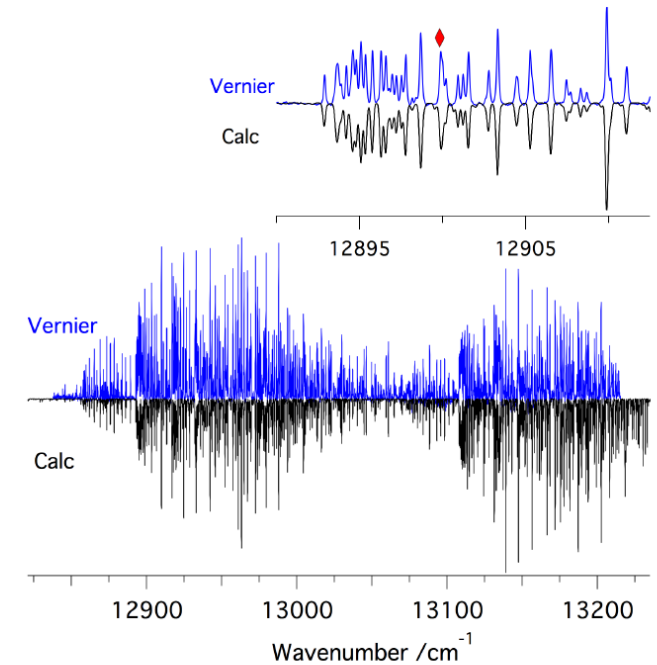
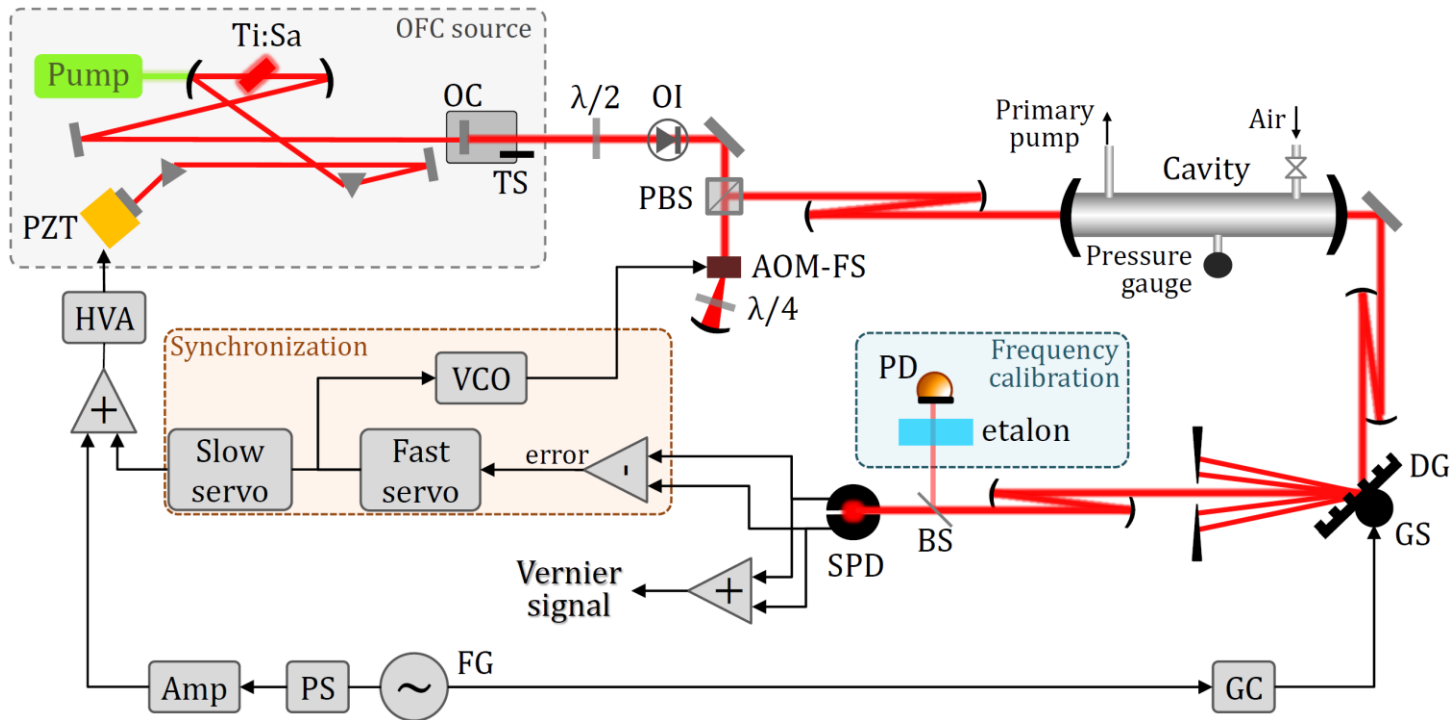


Figure 8. Comparison between Vernier spectrum (upper (blue) trace) and PGOPHER simulation of the $^{14}\text{N}_2$ $B-A$ (2-0) band (black trace). The discharge was operating with 0.5 Torr in pure N_2 , with 50 mA DC current; the simulation was performed setting $T = 380$ K, and a Gaussian FWHM of 0.18 cm^{-1} . The red diamond highlights an overlap of 3 lines (see Fig 7).

Journal of Quantitative Spectroscopy & Radiative Transfer 219 (2018) 127–141

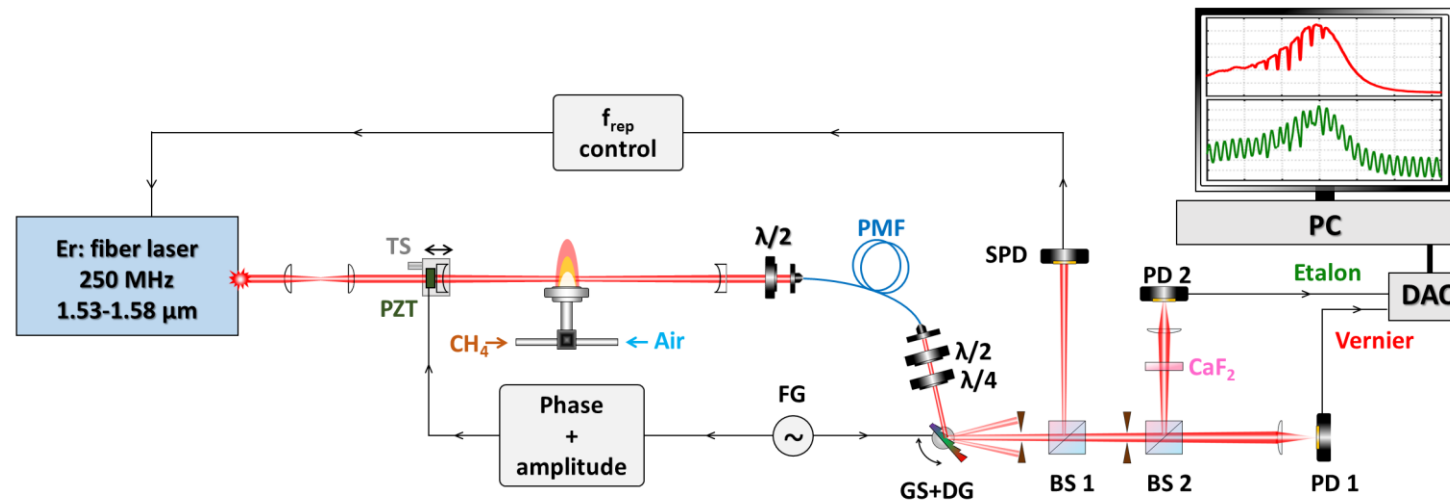
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Journal of Quantitative Spectroscopy & Radiative Transfer

journal homepage: www.elsevier.com/locate/jqsrt

NIR detection in a flame



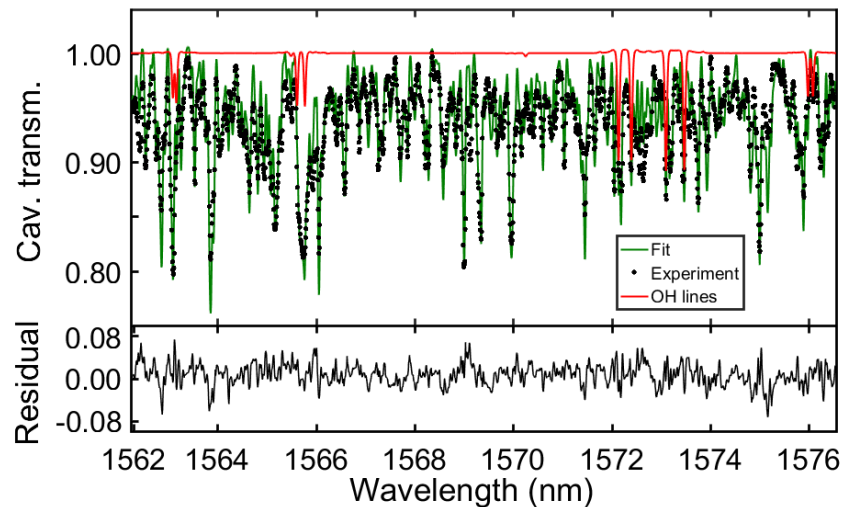
Flat flame burner*

- Flame diameter = 3.8 cm
- Flame temperature ~1950 K

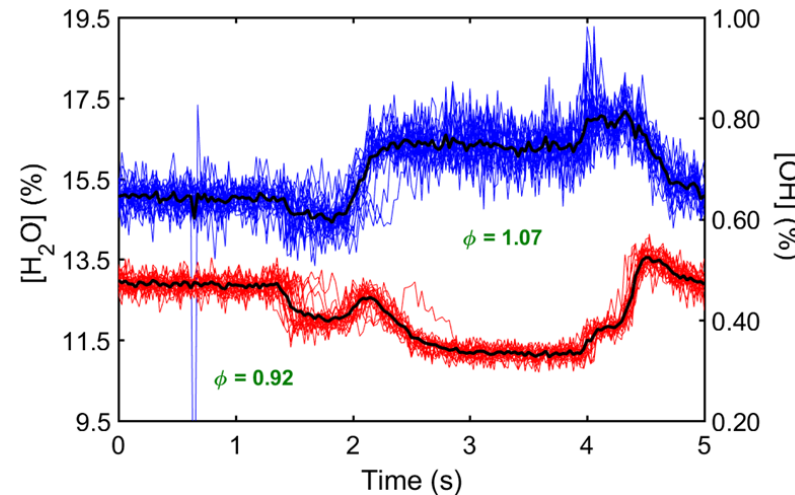
Measurements at stoichiometry

- CH₄ flow rate = 950 mL/min
- Air flow rate = 9050 mL/min

- Background spectrum recorded with flame off
- H₂O and OH radical transitions



[H₂O] and [OH] overlapped cycles and averages



Time-resolved continuous-filtering Vernier spectroscopy of H₂O and OH radical in a flame

CHUANG LU,^{1,3} FRANCISCO SENNA VIEIRA,^{1,3} FLORIAN M. SCHMIDT,² AND ALEKSANDRA FOLTYNOWICZ^{1,*}

¹Department of Physics, Umeå University, 901 87 Umeå, Sweden

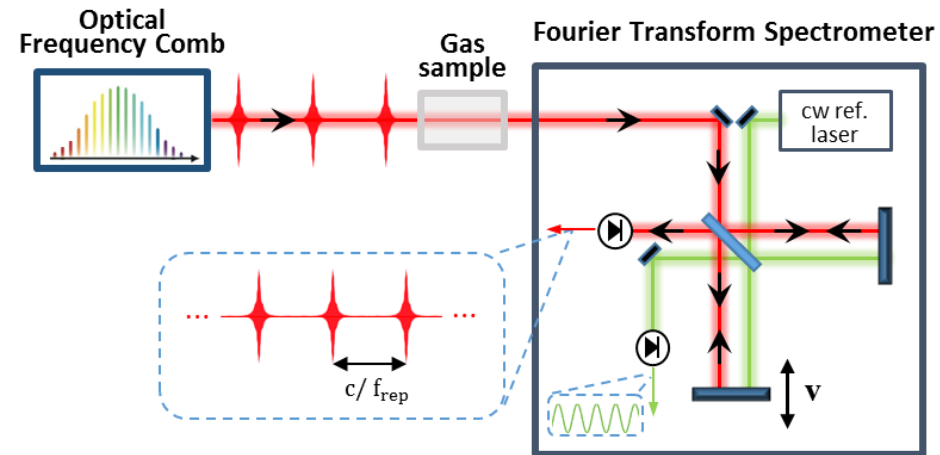
²Department of Applied Physics and Electronics, Umeå University, 901 87 Umeå, Sweden

Fourier transform approaches

Measurement in optical path difference (OPD)

- Interferogram + FFT
- Single detector
- Comb modes resolution possible
- Spectral coverage – entire comb range

Mechanical FTS Michelson interferometer



Fourier transform spectroscopy with a laser frequency comb

Julien Mandon, Guy Guelachvili and Nathalie Picqué*

Spectrochimica Acta Part A 75 (2010) 142–145



Contents lists available at ScienceDirect

Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy

journal homepage: www.elsevier.com/locate/saa



Demonstration of cavity enhanced FTIR spectroscopy using a femtosecond laser absorption source

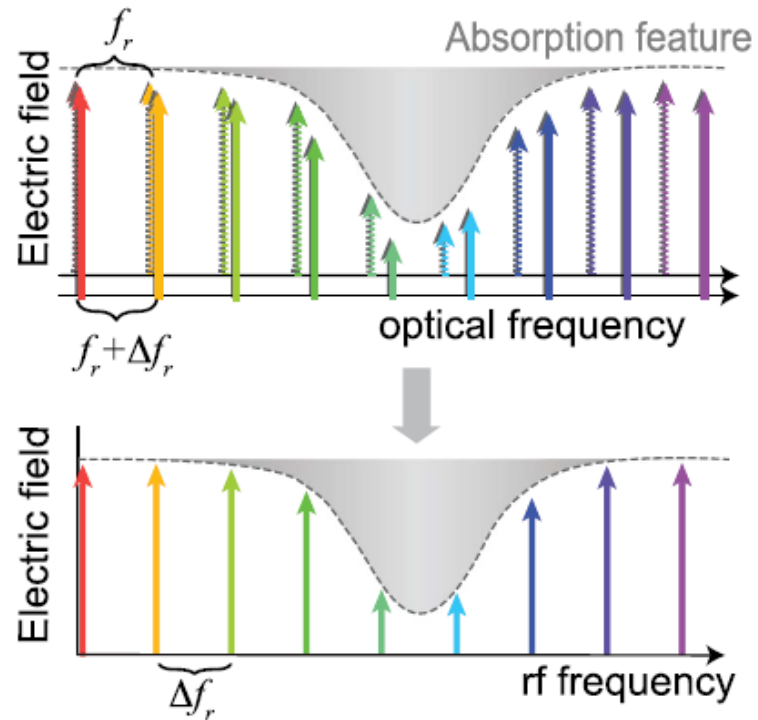
S. Kassi^a, K. Didriche^b, C. Lauzin^b, X. de Ghellinck d'Elseghem Vaernewijck^b, A. Rizopoulos^b, M. Herman^{b,*}

^a Laboratoire de Spectrométrie Physique (associated with CNRS, UMR 5588), Université Joseph Fourier de Grenoble, B.P. 87, 38402 Saint-Martin-d'Hères Cedex, France

^b Service Laboratoire de Chimie quantique et Photophysique, CP160/09, Faculté des Sciences, Université libre de Bruxelles (U.L.B.), Ave. Roosevelt 50, B-1050 Brussels, Belgium

Dual comb spectrometers

FTS without mechanical movement, instead two combs with different repetition rates



Time domain measurement

- Interferogram + abs(FFT)
- Single detector
- Comb modes resolution
- Spectral coverage – entire comb range
- Acquisition time – ms

Review Article

Vol. 3, No. 4 / April 2016 / Optica 414

optica

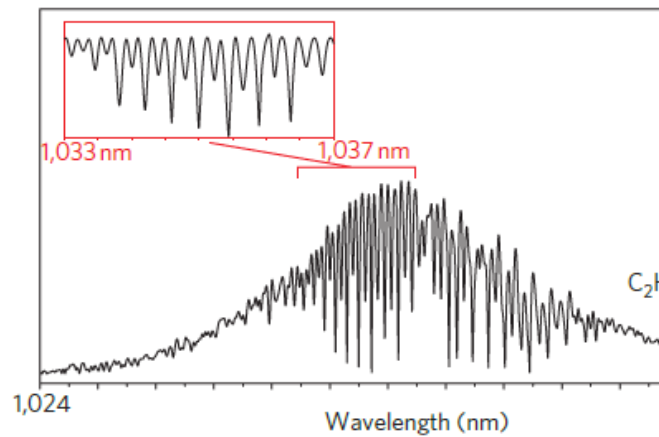
Dual-comb spectroscopy

IAN CODDINGTON,^{1,*} NATHAN NEWBURY,^{1,2} AND WILLIAM SWANN¹

¹National Institute of Standards and Technology, 325 Broadway, Boulder, Colorado 80305, USA

Cavity-enhanced dual-comb spectroscopy

Birgitta Bernhardt¹, Akira Ozawa¹, Patrick Jacquet², Marion Jacquey², Yohei Kobayashi³
Thomas Udem¹, Ronald Holzwarth^{1,4}, Guy Guelachvili², Theodor W. Hänsch¹



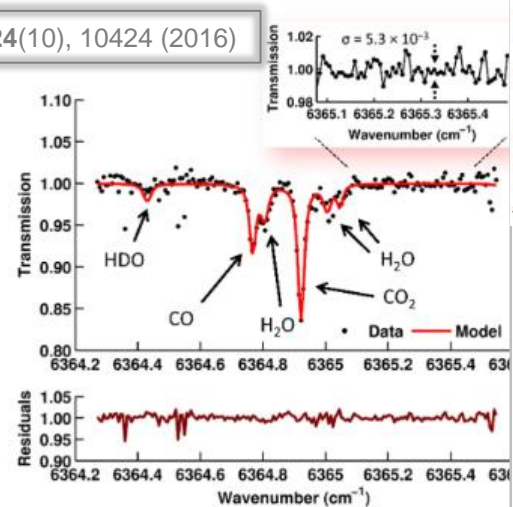
Coherent cavity-enhanced dual-comb spectroscopy

Adam J. Fleisher,^{1,*} David A. Long,^{1,3} Zachary D. Reed,¹ Joseph T. F. Plusquellic²

¹Material Measurement Laboratory, National Institute of Standards and Technology, Gaithersburg, Maryland, 20899, USA

²Physical Measurement Laboratory, National Institute of Standards and Technology, Colorado, 80305, USA

Opt. Express 24(10), 10424 (2016)



Research Article

Vol. 6, No. 1 / January 2019 / Optica 28

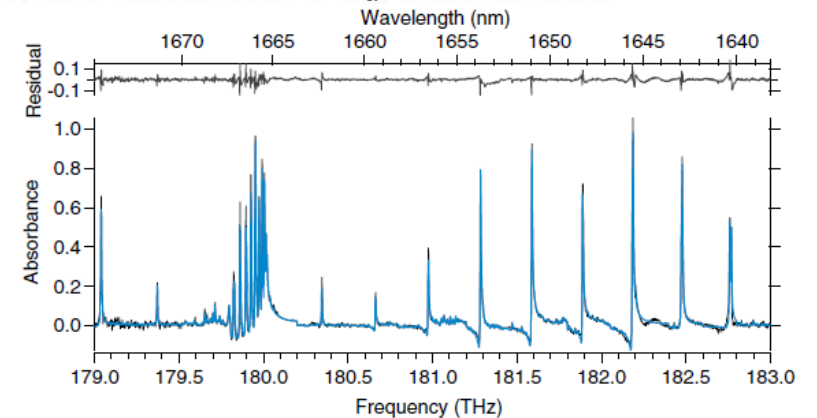


Broadband coherent cavity-enhanced dual-comb spectroscopy

NAZANIN HOGHOOGHI,^{1,*} ROBERT J. WRIGHT,¹ AMANDA S. MAKOWIECKI,¹ WILLIAM C. SWANN,²
ELEANOR M. WAXMAN,² IAN CODDINGTON,² AND GREGORY B. RIEKER¹

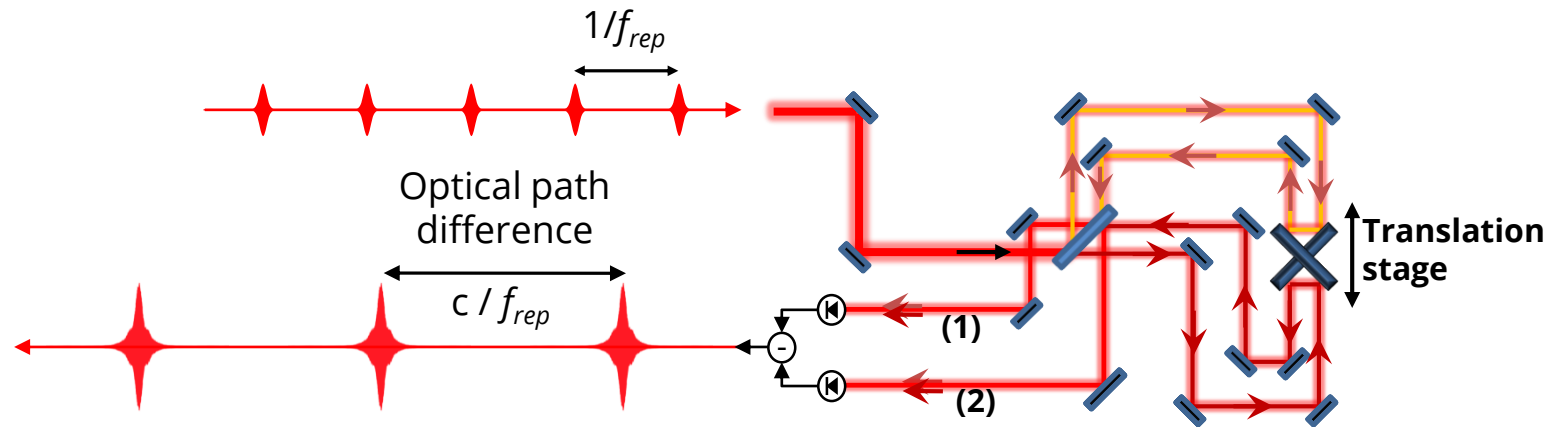
¹Precision Laser Diagnostics Laboratory, University of Colorado, Boulder, Colorado 80309, USA

²Applied Physics Division, National Institute of Standards and Technology, Boulder, Colorado 80305, USA



Mechanical FTS

Fast-scanning comb-based mechanical FTS



- Fast-scanning interferometer (1 m/s)
- 1 GHz resolution in 0.4 s
- Stabilized cw reference laser for OPD calibration

Auto-balanced detection

PRL 107, 233002 (2011)

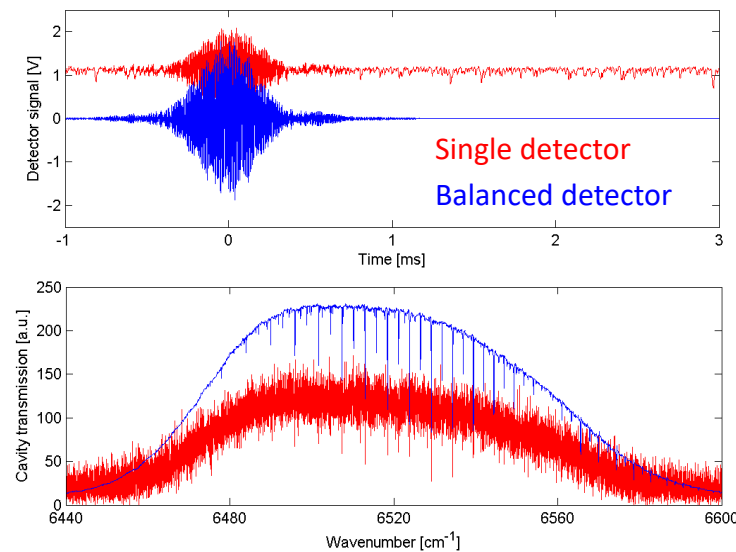
Selected for a **Viewpoint** in *Physics*
 PHYSICAL REVIEW LETTERS

week ending
 2 DECEMBER 2011



Quantum-Noise-Limited Optical Frequency Comb Spectroscopy

Aleksandra Foltynowicz,^{*} Tiejana Ban,[†] Piotr Masłowski,[‡] Florian Adler,[§] and Jun Ye
 JILA, National Institute of Standards and Technology and University of Colorado, Department of Physics,
 University of Colorado, Boulder, Colorado 80309-0440, USA



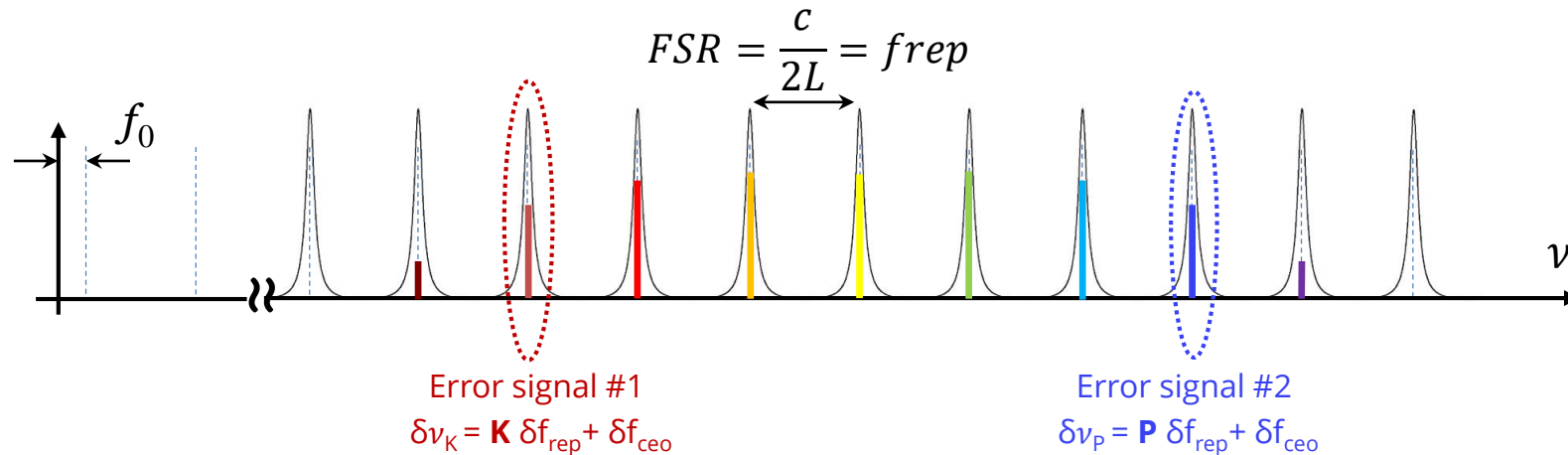
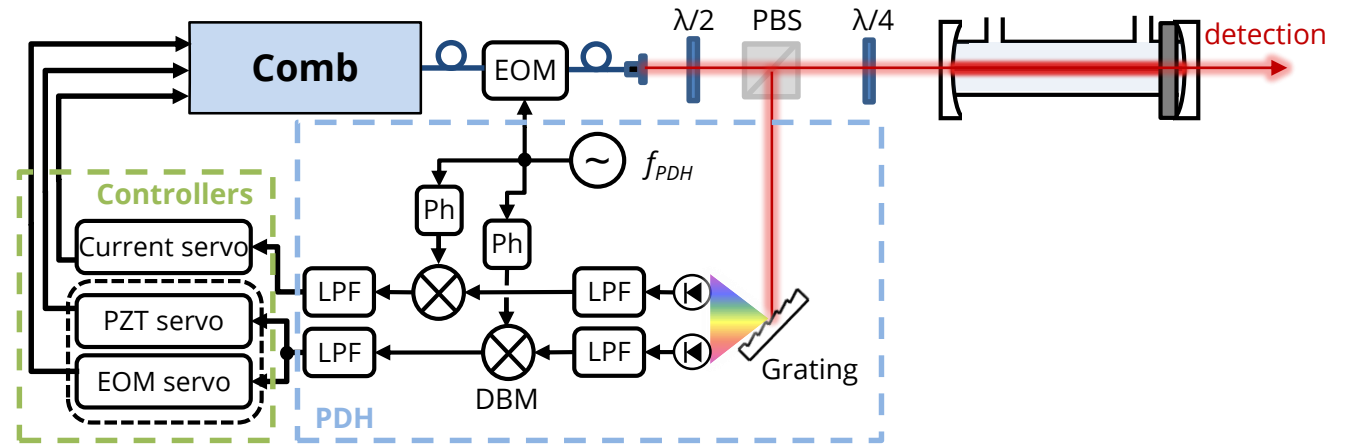


Comb-cavity coupling

Continuous transmission necessary:
2-point Pound-Drever-Hall locking technique

2nd locking point
Pump current ~150 kHz

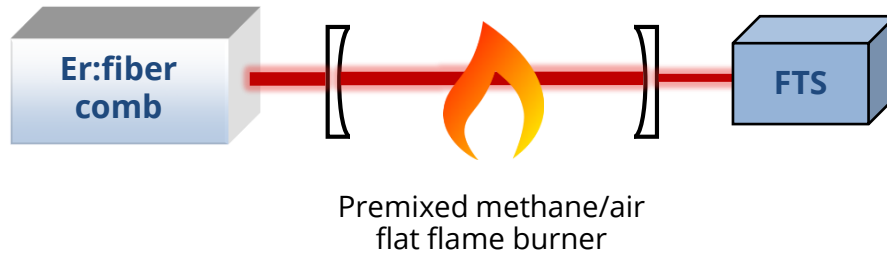
1st locking point
PZT ~6 kHz; EOM ~500 kHz



Transmitted bandwidth usually limited by the cavity dispersion



Detection in a flame

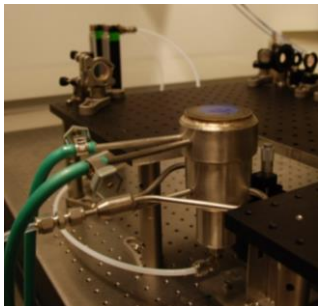


Cavity for flame measurement:

- finesse 150
- length 60 cm, FSR 250 MHz
- open air

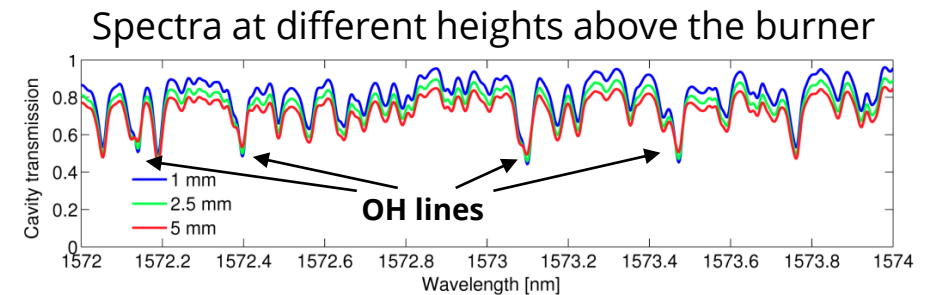
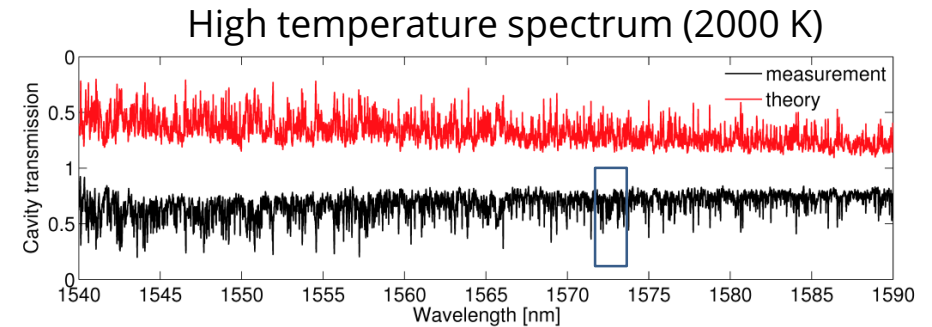
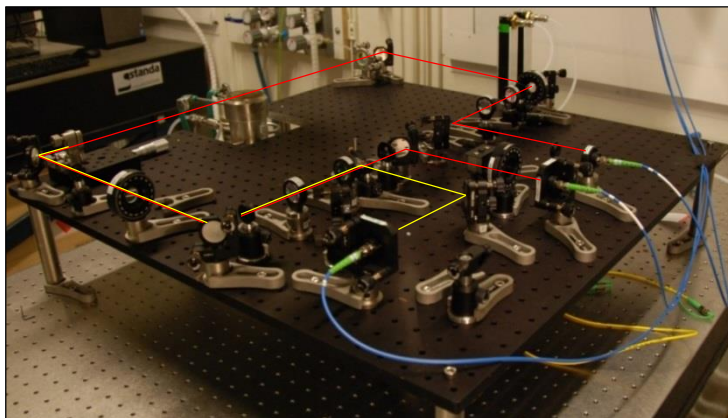
FTS:

- Resolution 1 GHz in 0.4 sec



High-temperature water and OH spectra in a flame

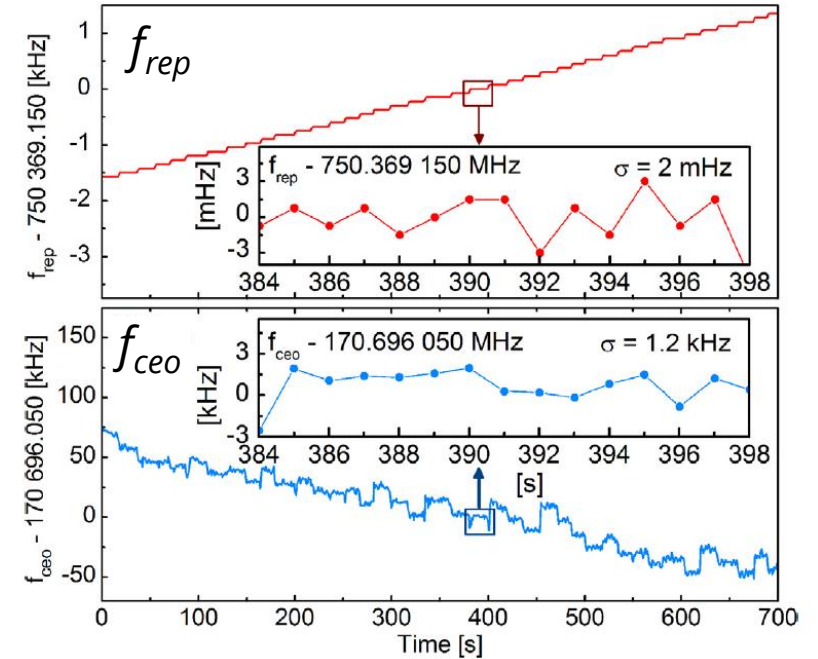
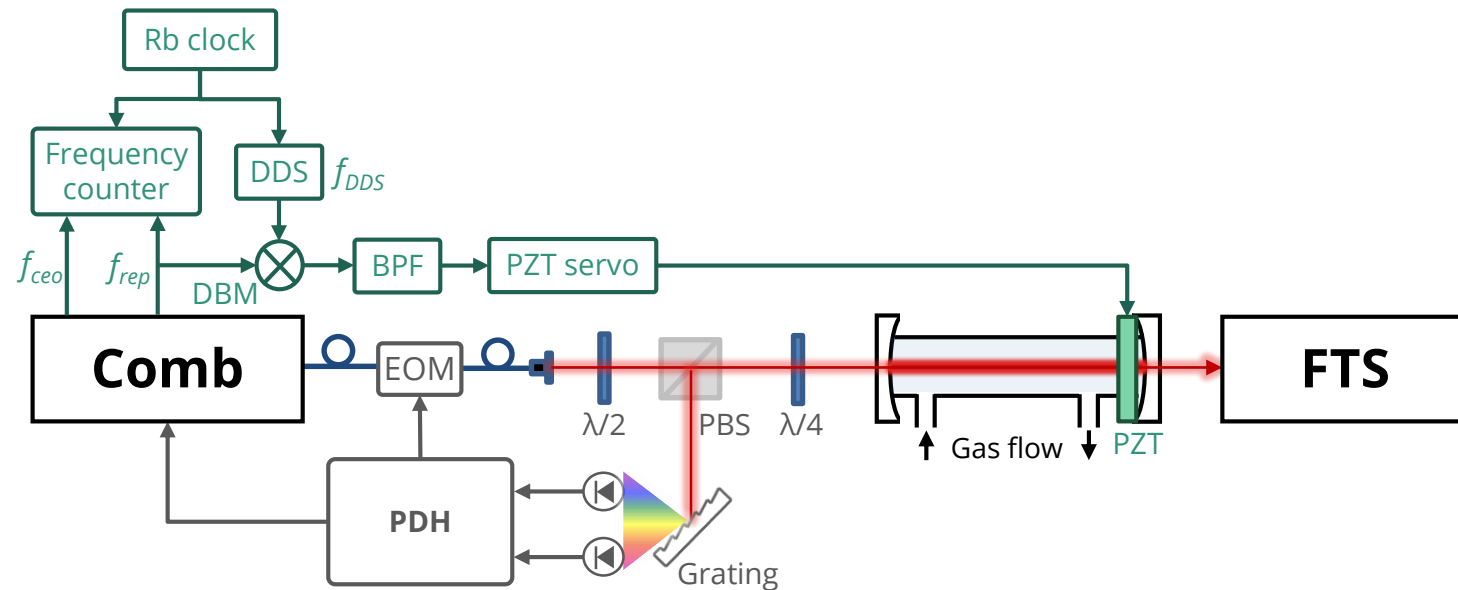
Remote operation



Absolute comb stabilization

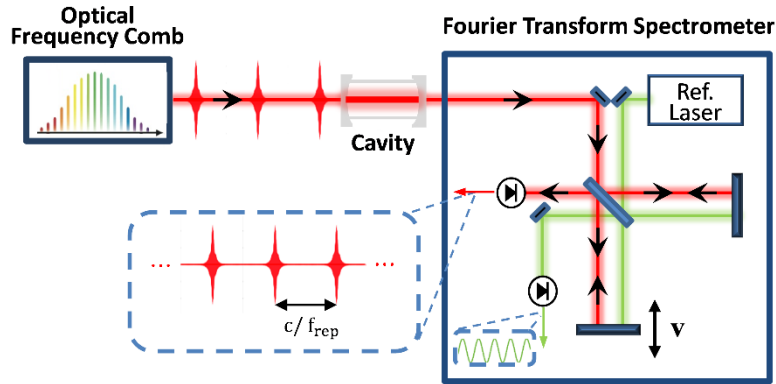
Stabilization of the absolute values of f_{rep} and f_{ceo}

- Require stabilization of the cavity length



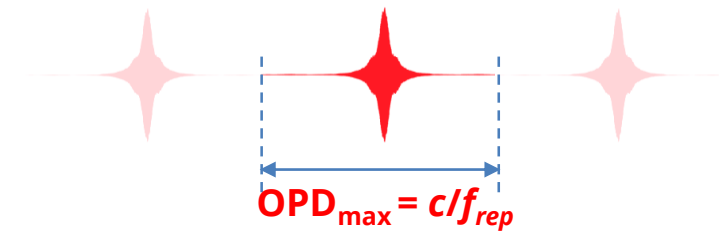


Sub-nominal resolution

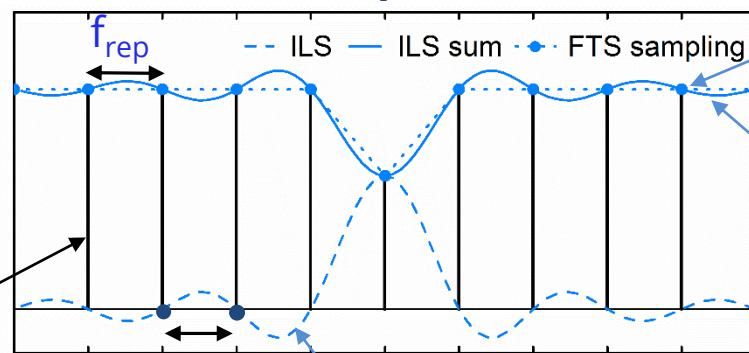


Train of bursts separated by c/f_{rep}

Single-burst interferogram
 Nominal resolution matched to f_{rep}
 Contains all spectral information



FTS spectrum



Comb line

FTS sampling points

FTS spectrum

Instrumental line shape (ILS)

PHYSICAL REVIEW A 93, 021802(R) (2016)

Surpassing the path-limited resolution of Fourier-transform spectrometry with frequency combs

Piotr Masłowski,¹ Kevin F. Lee,² Alexandra C. Johansson,³ Amir Khodabakhsh,³ Grzegorz Kowzan,¹ Lucile Rutkowski,³ Andrew A. Mills,² Christian Mohr,² Jie Jiang,² Martin E. Fermann,² and Aleksandra Foltynowicz^{3,*}

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Optical frequency comb Fourier transform spectrometry with sub-nominal resolution and precision beyond the Voigt profile

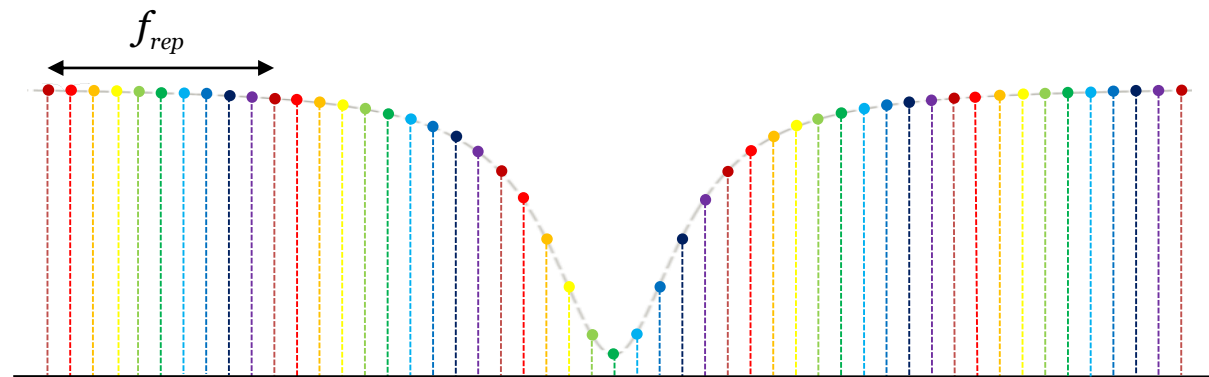
Lucile Rutkowski^a, Piotr Masłowski^b, Alexandra C. Johansson^a, Amir Khodabakhsh^a, Aleksandra Foltynowicz^{3,*}

^aDepartment of Physics, Umeå University, 901 87 Umeå, Sweden

^bInstitute of Physics, Faculty of Physics, Astronomy and Informatics, Nicolaus Copernicus University in Toruń, ul. Gruzdzka 5, 87-100 Toruń, Poland

Spectral interleaving

f_{rep} or f_{ceo} scan
to map the entire absorption lineshape

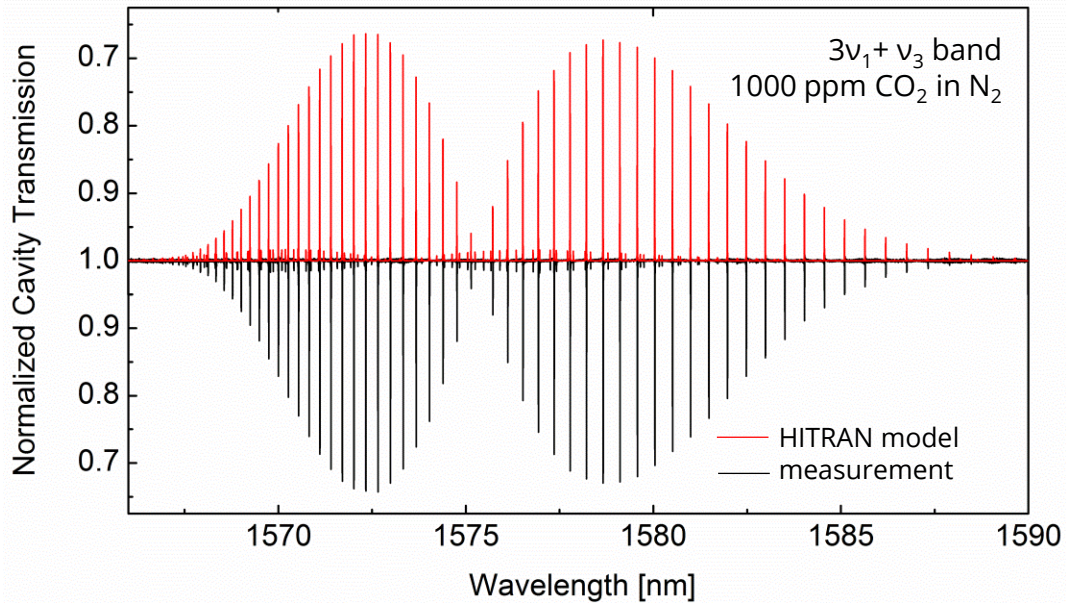


- Resolution – comb linewidth
- Frequency scale – given by the comb
- Sampling point spacing – arbitrary tuning
- Compact and fast spectrometer

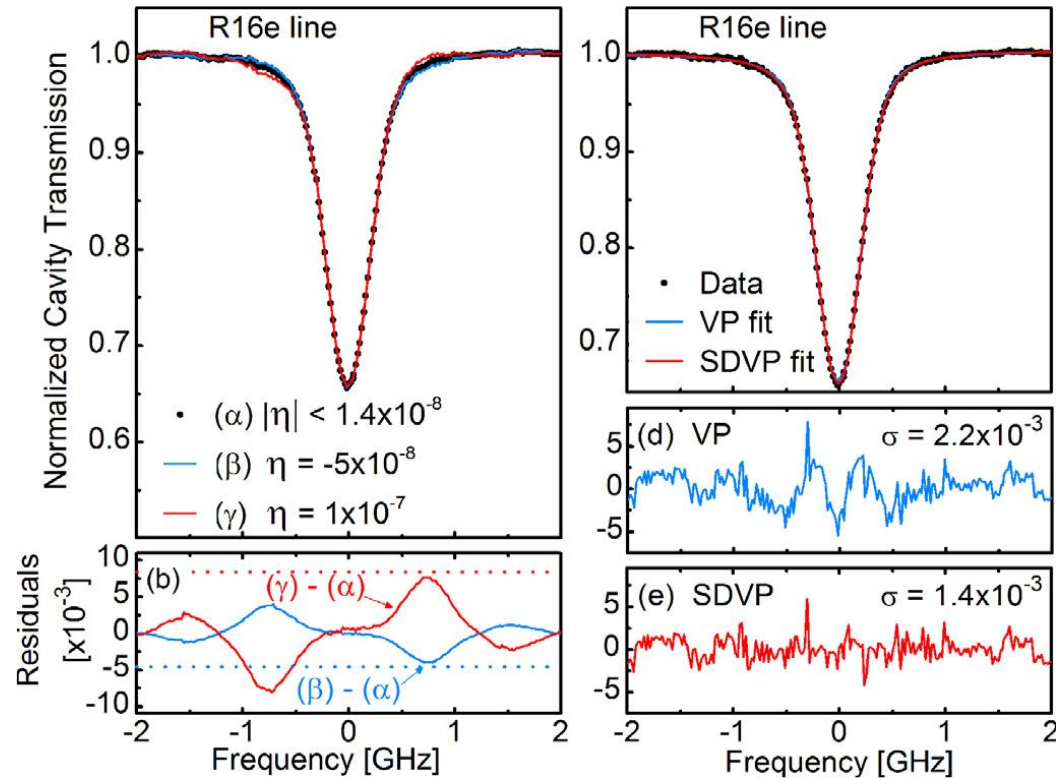
High precision spectroscopy



CO₂ in N₂ at 26 Torr



- Laser Er:fiber: 250 MHz
- Cavity - F = 10000, FSR = 187.5 MHz
- Interferogram: $\delta\nu_N = 750$ MHz (OPD_{max} = 40 cm)
- Optical step: 18.75 MHz
- 40 interleaved spectra
- Total acquisition time: 700 s



Optical frequency comb Fourier transform spectroscopy with sub-nominal resolution and precision beyond the Voigt profile

Lucile Rutkowski^a, Piotr Masłowski^b, Alexandra C. Johansson^a, Amir Khodabakhsh^a, Aleksandra Foltynowicz^{a,*}

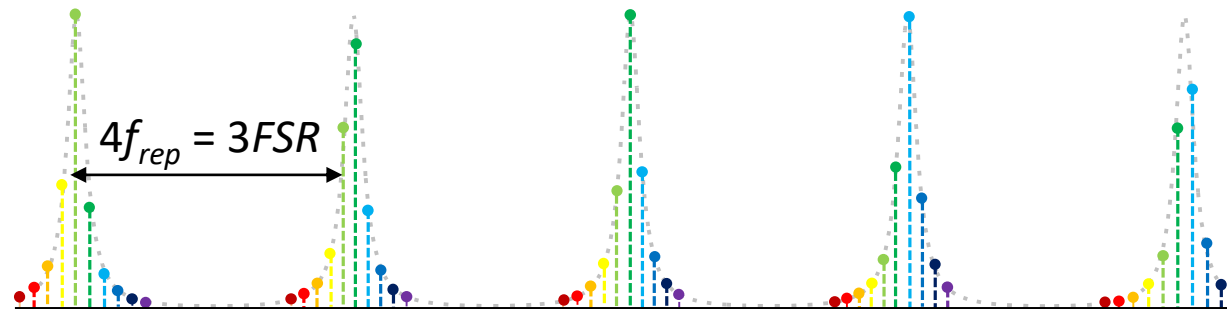
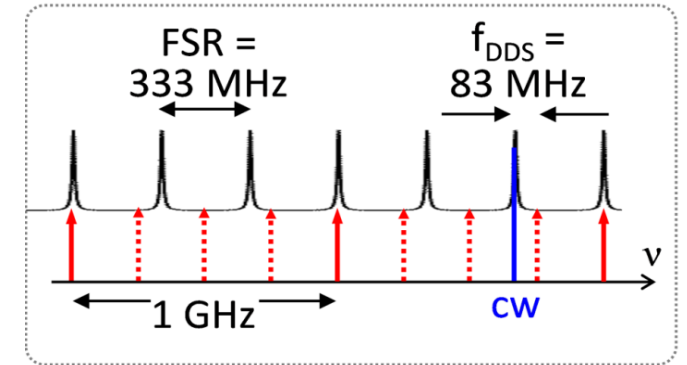
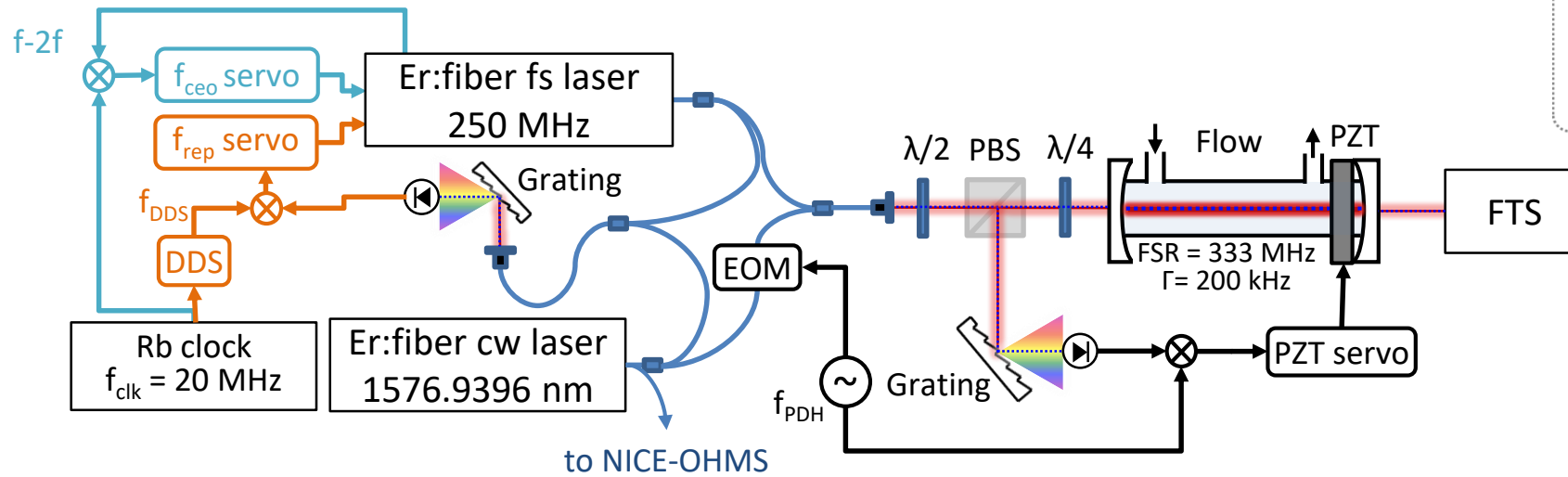
^aDepartment of Physics, Umeå University, 901 87 Umeå, Sweden

^bInstitute of Physics, Faculty of Physics, Astronomy and Informatics, Nicolaus Copernicus University in Toruń, ul. Gdazińska 5, 87-100 Toruń, Poland

Cavity mode spectroscopy



- Comb-cavity locking via a stabilized cw reference
- Cavity filled with 1% CO₂ in N₂ at 750 Torr

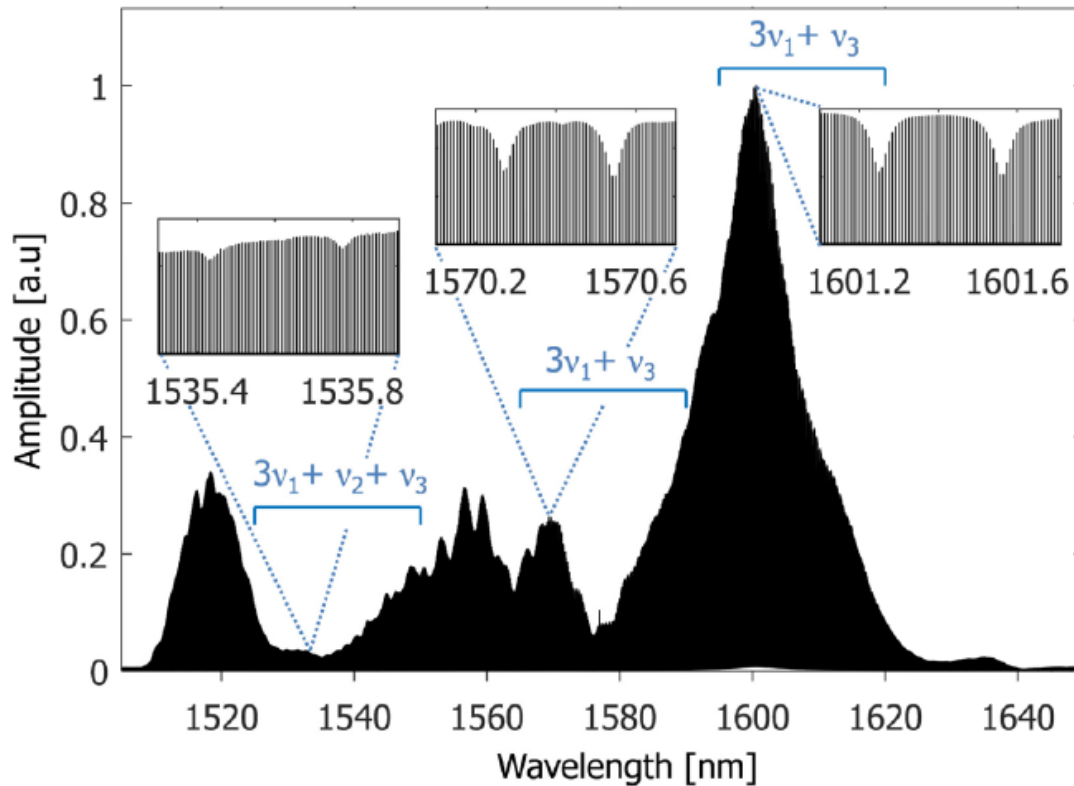


Cavity mode spectroscopy

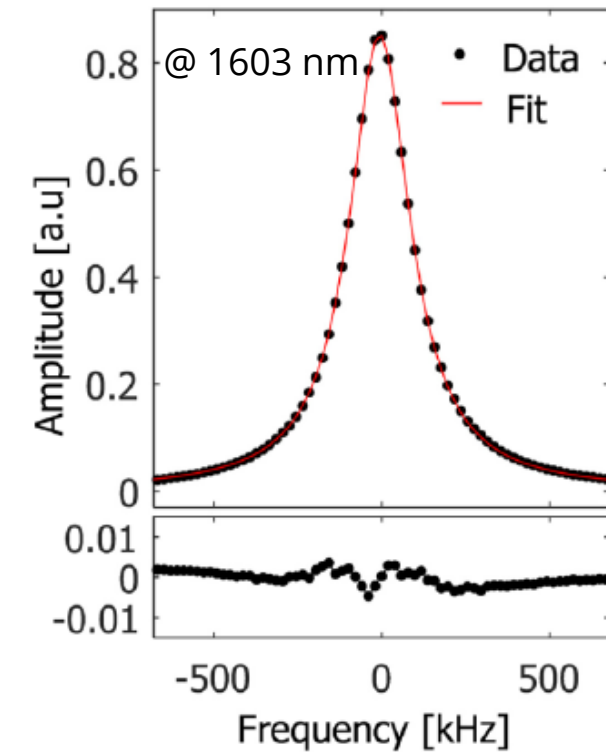


Full spectrum

Interleaving of 120 spectra taken with a 20 kHz optical step.



Lorentzian fit for each cavity mode:

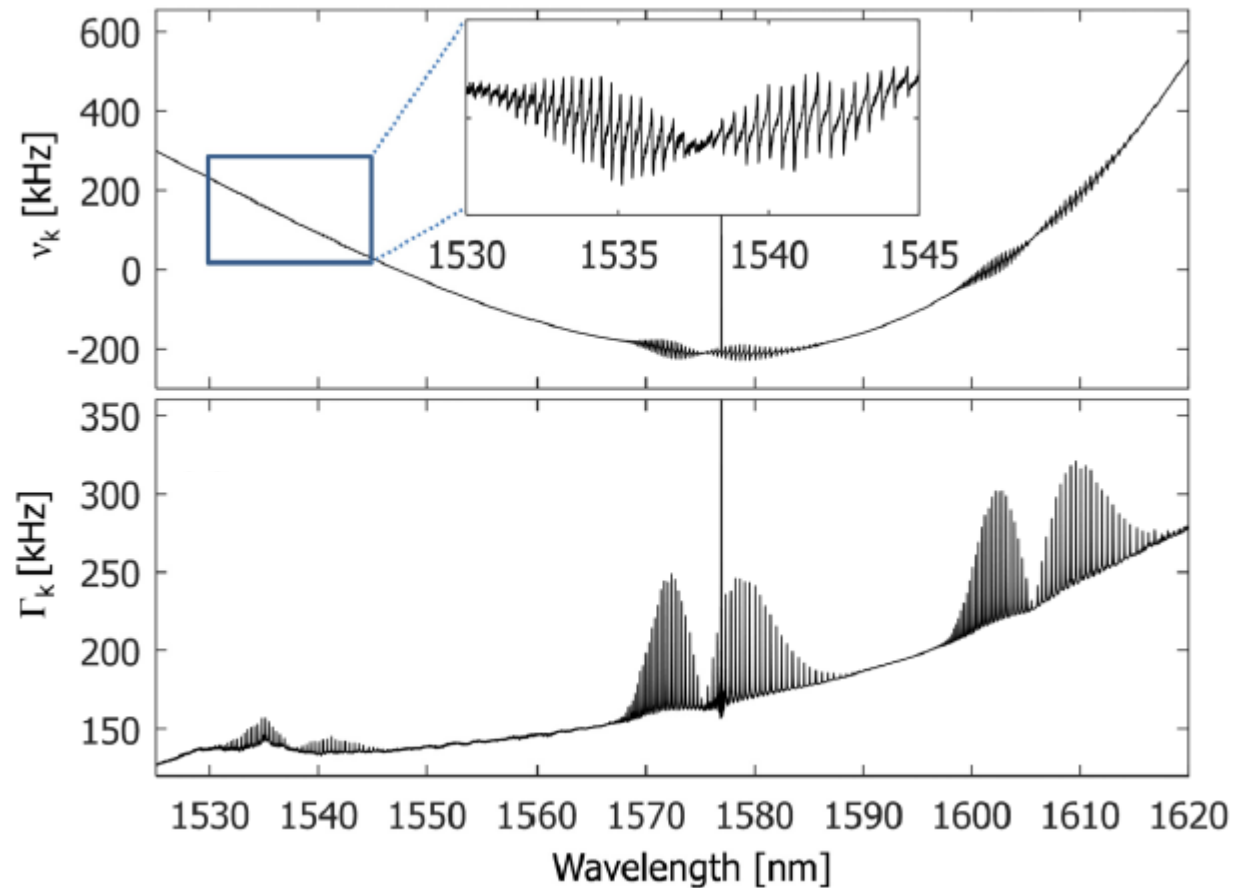


Cavity mode spectroscopy



Absorption and dispersion spectra from mode widths and center frequencies

Three absorption bands measured, no need to consider broadband dispersion effects.



Research Article

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Optics EXPRESS

Broadband calibration-free cavity-enhanced complex refractive index spectroscopy using a frequency comb

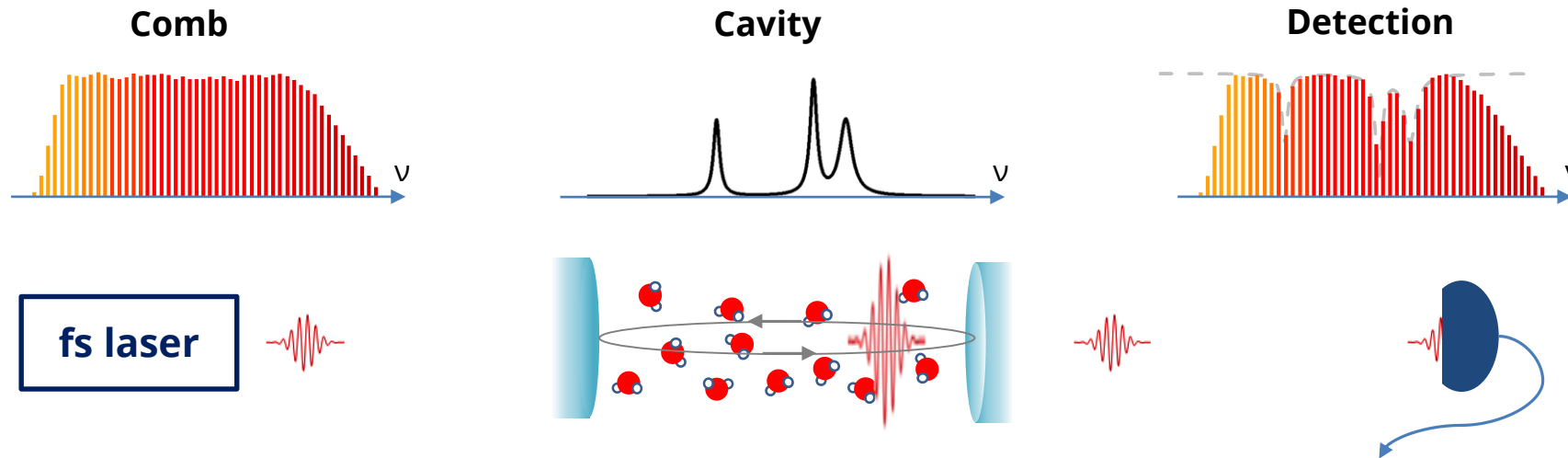
ALEXANDRA C. JOHANSSON,¹ LUCILE RUTKOWSKI,¹ ANNA FILIPSSON,¹
 THOMAS HAUSMANINGER,¹ GANG ZHAO,^{1,2} OVE AXNER,¹ AND
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²State Key Laboratory of Quantum Optics and Optics Devices, Institute of Laser Spectroscopy, Shanxi University, Taiyuan 030006, China

Conclusion

Measure entire absorption bands and/or different species **simultaneously**, with **high sensitivity** and in **short acquisition times**



Many techniques, balance between:

- Spectral coverage
- Spectral resolution
- Time resolution
- Sensitivity
- Total acquisition time
- Robustness

Vernier spectrometer in continuous filtering

- Use the enhancement cavity as a frequency filter
- Entire comb bandwidth, but loose the frequency ruler

Fourier transform spectroscopy

- Multiplex measurement, rely on a Michelson interferometer
- Easily combined with cavity enhancement, comb mode resolution



Review of Reviews

Optical frequency comb technology:

- N. R. Newbury, "Searching for applications for a fine tooth comb", Nat. Photon. **5**, 186 (2011).
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- M.L. Weichman, P.B. Changala, J. Ye, Z. Chen, M. Yan, N. Picqué, "Broadband molecular spectroscopy with optical frequency combs", J. Mol. Spectrosc. **355**, 66 (2019).

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- K. Twayana, I. Rebolledo-Salgado, E. Deriushkina, J. Schröder, M. Karlsson, V. Torres-Company, "Spectral Interferometry with frequency combs", Micromachines **13**(4), 614 (2022).

Application oriented

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- J.H. Lehman, M.L. Weichman, "Optical frequency comb for molecular spectroscopy, kinetics, and sensing", *Emerging Trends in Chemical Application of Lasers*, book chapter, 61-88 (2021).