BOOK OF ABSTRACTS

CES

Cavity Enhanced Spectroscopy

2022

June 14th-17th, Lecco, Italy

Contents

About

CES 2022 (Cavity Enhanced Spectroscopy)

CES (Cavity Enhanced Spectroscopy) is a biennial international meeting that brings together the community of physicists, chemists, atmospheric scientists, and engineers who work in the broad area of cavity enhanced spectroscopy and its many applications. The meeting is a forum for presentation of the latest developments in the field of optical cavities, including fundamental studies, micro- and nano-photonic themes, and applications in different areas of research. Investigators from academia, government and industry are encouraged to attend.

History

Lecco	2022	Greifswald	2007
Madison	2019	Cork	2006
Egmond aan Zee	2017	Oxford	2005
Boulder	2015	Eindhoven	2004
Naples	2013	Dusseldorf	2003
Kingston	2011	Lille	2002
Leiden	2009	Heeze	2001

Program Committee

Marco Marangoni	Politecnico di Milano	(Italy)
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Timetable

Tuesday, June 14th

14:00–14:10	CES2022 opening		
SUMMER SCHOOL	Chair:Marco Marangoni		
14:10–14:55	SS	**Adam J. Fleisher**	Accurate cavity-enhanced spectroscopy of molecules with coherent light
14:55–15:40	SS	**Lucile Rutkowski**	Cavity-enhanced optical frequency comb spectroscopy
15:40–16:25	SS	**Steven S. Brown**	Applications of cavity enhanced spectroscopy to atmospheric field measurements and aircraft research
16:25–16:45	Coffee break		
16:45–17:30	SS	**Oliver H. Heckl**	High finesse mirror design, fabrication and characterization
17:30–18.15	SS	**Randall Goldsmith**	Optical microcavities, a Low-Q Introduction

SS: Summer School Lecture

Wednesday, June 15th

	Chair:David Long		
9:00–9:35	IS	**Shui-Ming Hu**	Cavity-enhanced spectroscopy of molecules with sub-kHz accuracy
9:35–10:00	CT	**Szymon Wójtewicz**	Cavity-enhanced absorption and dispersion Doppler-free saturation spectroscopy
10:00–10:25	CT	**Daniele Romanini**	Comb coherence-transfer and high accuracy saturated cavity ring-down saturation spectroscopy
10:25–10:50	CT	**Adam J. Fleisher**	Resonance enhanced two-photon spectroscopy of nitrous oxide using a frequency-locked quantum cascade laser
10:50–11:10	Coffee break		
11:10–11:45	IS	**Ioachim Pupeza**	Cavity-enhanced field-resolved spectroscopy
11:45–12:10	CT	**Eckart Wrede**	A breath-by-breath, real-time acetone sensor based on cavity-enhanced laser-induced fluorescence
12:10–12:35	CT	**Davide Mazzotti**	Real-world applications of Saturated-Absorption CAvity Ring-down (SCAR) spectroscopy

12:35–13:00	CT	**Caroline Womack**	The miniature airborne cavity enhanced spectrometer (mACES) for sensitive NO_2 detection on UAV platforms
13:00–14:10	**Lunch**		
	Chair: Kevin Lehmann		
14:10–14:45	IS	**Peter Rakitzis**	Applications of cavity-based Polarimetry
14:45–15:10	CT	**Eugenio Fasci**	Comb-assisted cavity ring-down spectroscopy for ultra-sensitive traceable measurements of water vapour in ultra-high purity gases
15:10–15:35	CT	**Jean-Pierre van Helden**	The spatial distribution of species in an atmospheric pressure plasma jet investigated by cw cavity ring-down spectroscopy
15:35–16:00	CT	**Zhen Wang**	Ultra-high sensitivity and ultra-wide dynamic range gas detection based on photoacoustic spectroscopy
16:00–16:20	**Coffee break**		
16:20–16:55	IS	**Marissa Weichman**	Molecules in optical cavities: precision spectroscopy and strong light-matter interactions
16:55–17:20	CT	**Ibrahim Sadiek**	Mode-resolved Mid-Infrared optical frequency comb spectroscopy using an air-spaced VIPA without optical cavity filtering
17:20–17:45	CT	**Adrian Hjältén**	High-accuracy line positions of N_2O, CH_4 and H_2CO around 8 μm from optical frequency comb Fourier transform spectroscopy
17:45–17:55	SC	**Sven Schmidt**	NANOPLUS
17:55–18:05	SC	**Andreas Hugi**	IRSWEEP
19:00	**Happy Hour**		
20:00	**Poster Session**		

https://www.overleaf.com/project/629f69da929d193587a5ef12 *CT: Contributed Talk*
IS: Invited Speaker
SC: Sponsor Contribution

Thursday, June 16th

	Chair: Lucile Rutkowski		
9:00–9:35	IS	**Thomas Allison**	Understanding ultrafast observables via broadband cavity-enhanced ultrafast spectroscopy in jet-cooled molecules and clusters
9:35–10:00	CT	**Romain Dubroeucq**	High finesse Fourier transform cavity ring-down spectroscopy with an optical frequency comb
10:00–10:25	CT	**Dominik Charczun**	Electro-optical dual-comb cavity ring-down, mode-width and mode-dispersion spectroscopy

10:25–10:50	CT	**Vinicius Silva de Oliveira**	Cavity-enhanced double-resonance spectroscopy of methane using a frequency comb probe
10:50–11:10	**Coffee break**		
	Chair: Carlos Saavedra		
11:10–11:45	IS	**Silvia Soria Huguet**	Microbubble resonators as enhanced platforms for thermometry and flow cytometry
11:45–12:10	CT	**David Long**	Optomechanical devices for high accuracy acceleration sensing
12:10–12:35	CT	**Marcin Makowski**	High-finesse optical cavity length adjustment at cryogenic temperatures
12:35–13:00	**Round table: Designing the new CES meeting**		
13:00–14:10	**Lunch**		
	Chair Oliver H. Heckl		
14:10–14:45	IS	**Marco Lamperti**	Cavity-enhanced vs coherent Raman metrology of H_2
14:45–15:10	CT	**Philip Martin**	Infrared off-axis cavity-enhanced absorption spectroscopy of molecular hydrogen
15:10–15:35	CT	**Daniele Romanini**	Fast breath analysis by OF-CEAS: clinical studies for lung transplantation
16:00	**Touristic excursion: Bellagio and Villa Melzi d'Eril**		
20:00	**Gala dinner**		

CT: Contributed Talk
IS: Invited Speaker

Friday, June 17th

	Chair: Hans-Peter Loock		
9:00–9:35	IS	**Andreas Reiserer**	Spectroscopy and control of individual erbium dopants in optical resonators
9:35–10:00	CT	**Maximilian Högner**	Broadband nonlinear cavity ring-down spectroscopy
10:00–10:25	CT	**Shalom Palkhivala**	Microcavity-enhanced investigation of nanoparticle dynamics
10:25–10:50	CT	**Nikita Toropov**	Single-molecule thermo-optoplasmonic sensing of enzymes
10:50–11:10	**Coffee break**		
	Chair: Randall Goldsmith		
11:10–11:45	IS	**Carlos Saavedra**	Fiber Fabry-Pérot micro-resonators for molecular oxygen A-band determination using modulation spectroscopy
11:45–12:10	CT	**Tao Lu**	A silica rib microdisk for visible comb spectroscopy

12:10–12:35	CT	**Oliver Hirschmann**	Adding quantum confinement to cavity molecular polaritons
12:35–13:00	CT	**Hans-Peter Loock**	Fiber optic strain sensing with pi-shifted fiber Bragg gratings
13:00–13:10	**Greetings and closing remarks**		
13:10–14:10	**Lunch**		

CT: Contributed Talk
IS: Invited Speaker

List of Abstracts – Talks

Tuesday, June 14th

Accurate cavity-enhanced spectroscopy of molecules with coherent light

Adam J. Fleisher SS

National Institute of Standards and Technology (NIST), Boulder, CO, USA

Accurate spectral reference data for molecules unpins numerous sensing applications, including the remote sensing of atmosphere and trace gas detection. To accurately measure spectral reference data, we require precision laboratory techniques to be performed under well-controlled sample conditions. This summer school presentation will introduce several precise and accurate laboratory measurement techniques that leverage the advantages of cavity-enhanced spectroscopy which include high sensitivity, certain noise immunities, and multiplexed acquisition. Working examples of accurate spectroscopy and its application to challenges like traceable isotope analysis [1] will be discussed.

References

[1] A. J. Fleisher, H. Yi, A. Srivastava, O. L. Polyansky, N. F. Zobov, and J. T. Hodges, "Absolute $^{13}C/^{12}C$ isotope amount ratio for Vienna PeeDee Belemnite from infrared absorption spectroscopy", Nature Physics **17**, 889–893 (2021).

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Cavity-enhanced optical frequency comb spectroscopy

Lucile Rutkowski SS

University of Rennes, CNRS, Institut de Physique de Rennes (IPR), Rennes, France

Cavity-enhanced spectroscopy based on optical frequency comb sources combines sensitivity, spectral coverage and high resolution with limited sample volumes and short acquisition times. Since 2002, many developments have pushed the performances of such spectrometers forward, making them suitable for many different types of applications. I will give an overview of the different existing approaches before focusing on the Vernier and mechanical Fourier transform spectrometers. These last techniques have now reached maturity and found multiple applications ranging from high precision spectroscopy to applied measurements.

Applications of cavity enhanced spectroscopy to atmospheric field measurements and aircraft research

Steven S. Brown SS

National Oceanic and Atmospheric Administration (NOAA) Chemical Sciences Laboratory, Boulder, CO, USA

Cavity Enhanced Spectroscopy (CES) is now a widely used analytical method for measurements of atmospheric trace gases and extinction due to aerosol particles and Rayleigh scattering. Its advantage over more traditional absorption spectroscopy methods is high sensitivity at the level required in atmospheric science, which is typically in the optical extinction range of $10^{-11} - 10^{-7}$ cm^{-1}. As a form of direct absorption spectroscopy, CES has the further advantage of being an absolute analytical method requiring less frequent or less cumbersome calibration approaches than other analytical approaches. Techniques such as fluorescence spectroscopy or mass spectrometry are often higher in sensitivity for trace gas or aerosol composition measurements, but require calibration of the instrument response function. Indeed, CES is now often used both as a direct instrumental method and as a reference for other analytical techniques. Spectroscopically based instruments also offer considerable potential for miniaturization, especially relative to methods that require high vacuum. This summer school lecture will survey different approaches to CES commonly used in atmospheric science, including cavity ring-down spectroscopy (CRDS), integrated cavity output spectroscopy (ICOS), incoherent broadband cavity enhanced absorption (extinction) spectroscopy (IBBCEAS), and cavity attenuated phase shift spectroscopy (CAPS). It will also survey the utility of different spectral regions and light sources from the visible to the mid infrared. Examples of different instruments will include both custom built and commercially available systems. The NOAA Chemical Sciences Laboratory in Boulder, Colorado has a strong field program in atmospheric chemistry research that provides numerous examples of applications of CES for atmospheric trace gas sensing and aerosol extinction. The program has developed custom instruments from scratch and utilized custom-modified commercial instruments. Examples for this presentation will include measurements from ground sites, tall towers, ships and research aircraft, with an emphasis on the latter.

High finesse mirror design, fabrication and characterization

Oliver H. Heckl SS

University of Vienna, Vienna, Austria

High performance mirrors are employed for the construction of optical resonators in a variety of applications in optics and photonics. Stable resonators are routinely used to narrow the linewidth of continuous-wave lasers, thereby creating optical references for frequency comb stabilization and precision molecular spectroscopy [1–3]. Stable interferometers can be very small or very large in size, enabling scientific discovery in fields as seemingly unrelated as microcavity sensing [4] and gravitational wave detection [5]. Emerging applications in chemical sensing [6], discrete imaging [7], ultracold chemistry [8, 9], and even fundamental physics [10] benefit from high-performance mirrors.
During this summer school lecture, we look into the function principle of high-performance mirrors based on distributed Bragg reflectors, their design and design constraints, fabrication methods, spectral coverage and its limitations towards the UV and mid-IR spectral ranges. Before concluding, we will also discuss measurement schemes that allow mirror characterization on the ppm-level [11].

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Optical Microcavities, a Low-Q Introduction

Randall Goldsmith

SS

Department of Chemistry, University of Wisconsin, Madison, USA

The basic workings and operating equations of optical cavities will be presented from a microcavity perspective. Examples will be taken from recent microcavity literature with applications in chemical sensing, optomechanics, and quantum information science.

Wednesday, June 15th

Cavity-enhanced spectroscopy of molecules with sub-kHz accuracy

Shui-Ming Hu IS

University of Science and Technology of China, Hefei, China

Precise determination of ro-vibrational transition frequencies of molecules is interested in metrology as well as fundamental physics. The use of high-finesse optical cavities not only enhances the detection sensitivity but also provides a strong laser field that may saturate weak overtone transitions of molecules. Comb-locked cavity-enhanced absorption spectroscopy instruments were developed to measure Doppler-free spectroscopy of molecules. Two-color double resonance spectroscopy with milli-watt diode lasers was also established. We demonstrate a detection sensitivity of $10^{-12}/$cm together with sub-kHz frequency precision. Several applications will be presented, including abnormal line shift and broadening due to collisions at low pressures, the saturated spectroscopy of HD towards a determination of the proton-to-electron mass ratio, and detection of trace molecules/isotopes with cavity-enhanced double resonance spectroscopy.

Cavity-enhanced absorption and dispersion Doppler-free saturation spectroscopy

Szymon Wójtewicz, Katarzyna Bielska, Jolanta Domysławska, Aleksandr Balashov, MichałSłowiński, Marcin Bober, Agata Cygan, Magdalena Konefał, Grzegorz Kowzan, Mikołaj Zaborowski, Dominik Charczun, Piotr Wcisło, Piotr Masłowski, Roman Ciuryło, Daniel Lisak IS

Institute of Astronomy, Faculty of Physics, Astronomy and Informatics, Nicolaus Copernicus University, Toruń, Poland

Measurements of the absolute frequency of atomic and molecular transitions are the most sensitive tool for fundamental studies and many applications like remote sensing or optical metrology. Here we present an accurate determination of the positions of several molecular transitions using the Doppler-free saturated-absorption or dispersion spectroscopy. Our targets are near-infrared CO 3-0 band and CO_2 $(30012) \longleftarrow (00001)$ band as well as visible O_2 B band. We applied three ultra-sensitive cavity-enhanced techniques: the well-established cavity ring-down spectroscopy (CRDS), the cavity mode-width spectroscopy (CMWS) [1], and emerging cavity mode-dispersion spectroscopy (CMDS) [2]. The last two techniques were employed for the first time for the Lamb dip spectroscopy [3]. The CMDS method is advantageous to the other two: it enables measurements in broader pressure range and shows high immunity of the Lamb dip position to the simplifications in the model of saturated cavity mode shape as well as saturating power variations. The unperturbed transition frequencies are determined with even sub-kHz standard uncertainties, translating to relative uncertainties below $3 \cdot 10^{-12}$. The results are compared with the previous data [4–7].

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Comb coherence-transfer and high accuracy saturated cavity ring-down saturation spectroscopy

Daniele Romanini[1], ***Ondrej Votava***[2], ***Samir Kassi***[1], ***Alain Campargue***[1]

CT

[1]Université Grenoble Alpes and CNRS, Grenoble, France
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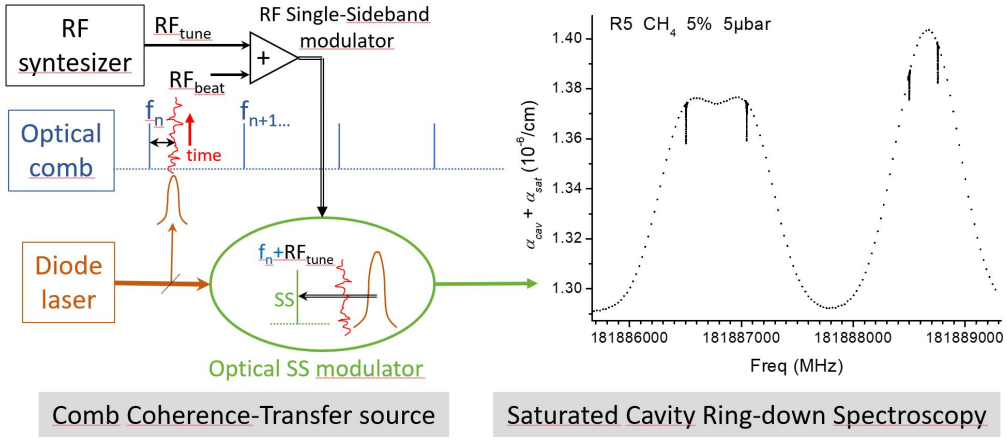
Comb Coherence Transfer (CCT) exploits a feed-forward frequency correction scheme to transfer the optical phase of a frequency comb to the emitted radiation of a free-running DFB diode laser. This allows to amplify by 4 or 5 orders of magnitude a selected comb tooth, while adding agile and accurate frequency tuning. In our laboratory, SI-traceable frequency calibration and comb tooth narrowing down to 20 kHz is provided by comb frequency locking to an ultrastable optical frequency reference distributed from Paris to Grenoble through the RENATER optical fiber network [1]. We applied this CCT broadly tunable source for saturated cavity ring-down



Comb Coherence-Transfer source | Saturated Cavity Ring-down Spectroscopy

spectroscopy of ro-vibrational multiplets in the $2\nu_3$ band of $^{12}CH_4$ from 6015 to 6115 cm^{-1}. Efficient cavity injection with large intra-cavity power build-up induces saturation of these transitions at low pressure, thus Doppler-free Lamb dips are observed, with high signal/noise. kHz-accurate transition frequencies are obtained [2], improving by three orders of magnitude previous values from spectra in the Doppler regime, which are strongly affected by line blending. While previous saturation spectroscopy investigations addressed specific $2\nu_3$ multiplets (R6 or R9), the CCT approach allowed for a rapid coverage of the entire R0-R10 series. Measured transition frequencies are compared with experimental and theoretical line lists available in the literature.

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Resonance enhanced two-photon spectroscopy of nitrous oxide using a frequency-locked quantum cascade laser

Gang Zhao[1]**,** ***Jianfei Tian***[2]**,** ***D. Michelle Bailey***[1]**,** ***Joseph T. Hodges***[1]**,** ***Kevin K. Lehmann***[3]**,** ***Adam J. Fleisher***[1]

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[3]Departments of Chemistry and Physics, University of Virginia, Charlottesville, VA, USA

We report the Doppler-free two-photon absorption of nitrous oxide (N_2O), observed at a wavelength of 4.53 μm. This resonance enhanced two-photon transition within the ν_3 vibrational overtone band of N_2O — measured by both cavity ring-down spectroscopy and cavity-enhanced absorption spectroscopy — is shown to be a sensitive and selective probe for trace molecules in a gas sample, as predicted by Lehmann [1].
To probe the Doppler-free transition with high frequency resolution, we demonstrate frequency locking of a quantum cascade laser (QCL) by weak resonant feedback from a two-mirror, high finesse optical resonator which also acts as the gas sample cell. We present a steady-state model for optical feedback from the Fabry-Perot cavity which, importantly, accounts for the difference in phase between the resonant (leak-out) and non-resonant (prompt reflection) electric fields returning to the QCL probe. Frequency noise analysis of the free-running and locked lasers shows a dramatic reduction in laser linewidth which enabled Doppler-free two-photon spectroscopy of N_2O.

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Cavity-enhanced field-resolved spectroscopy

Philipp Sulzer[1], ***Maximilian Högner***[1], ***Ann-Kathrin Raab***[1], ***Lukas Fürst***[2], ***Ernst Fill***[1], ***Daniel Gerz***[2], ***Christina Hofer***[2], ***Liudmila Voronina***[1], ***Ioachim Pupeza***[1] **IS**

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Femtosecond enhancement cavities (fsEC) are key to applications including high-sensitivity linear and nonlinear spectroscopy, as well as efficient nonlinear optical frequency conversion. Broadband passive enhancement in an fsEC, however, imposes the challenging requirements to the mirrors of high reflectivity and flat spectral phase over the entire bandwidth. As a consequence, to date, the broadest simultaneously-enhanced bandwidths amount to $< 20\%$ of the central optical frequency. Furthermore, so far, fsECs have covered only fractions of the molecular fingerprint region.
In this contribution, we present an ultrabroadband fsEC comprising gold-coated mirrors and a reflective wedged-diamond-plate input coupler, with an average finesse of 55 for optical frequencies below 40 THz ($\lambda > 7.50$ μm), and exceeding 40 in the 120-THz to 300-THz (1 to 2.5 μm) range. By controlling the carrier-envelope phase acquired upon each roundtrip by means of varying the atmospheric humidity, we demonstrate the simultaneous enhancement of an offset-free 50 MHz-repetition-rate frequency comb, obtained by intrapulse difference-frequency generation, in the 22-to-40-THz range. This results in an ultrashort circulating pulse with fixed carrier-envelope phase and with a Fourier limit of 1.6 cycles and a central wavelength of 9.5 μm. The pulse-to-pulse waveform-stable electric field of this offset-free comb enables time-domain electric-field-resolved detection of the excitation pulse and of the waves at the fsEC reflection and transmission ports via free-space electro-optic sampling (EOS).
The talk will address the advantages of combining this ultrabroadband fsEC with field-resolving detection. Firstly, unlike in time-integrating spectroscopies, EOS enables the detection of resonant molecular emission in the wake of an impulsive excitation in the absence of background from the latter. The contrast between the two is enhanced by the fsEC by taking advantage of destructive interference at the input coupler, occurring in the reflection port. Secondly, with recent advances in in EOS sensitivity, the effective interaction length with an intracavity gas of up to 81 m results in the prospect of ppt-level sensitivity for broadband infrared vibrational spectroscopy. Finally, the power enhancement exceeding a factor of 10, along with the capability of enhancing single few-cycle pulses make this fsEC design highly interesting for driving and investigating nonlinear processes in the long-wave infrared spectral range.

A breath-by-breath, real-time acetone sensor based on cavity-enhanced laser-induced fluorescence

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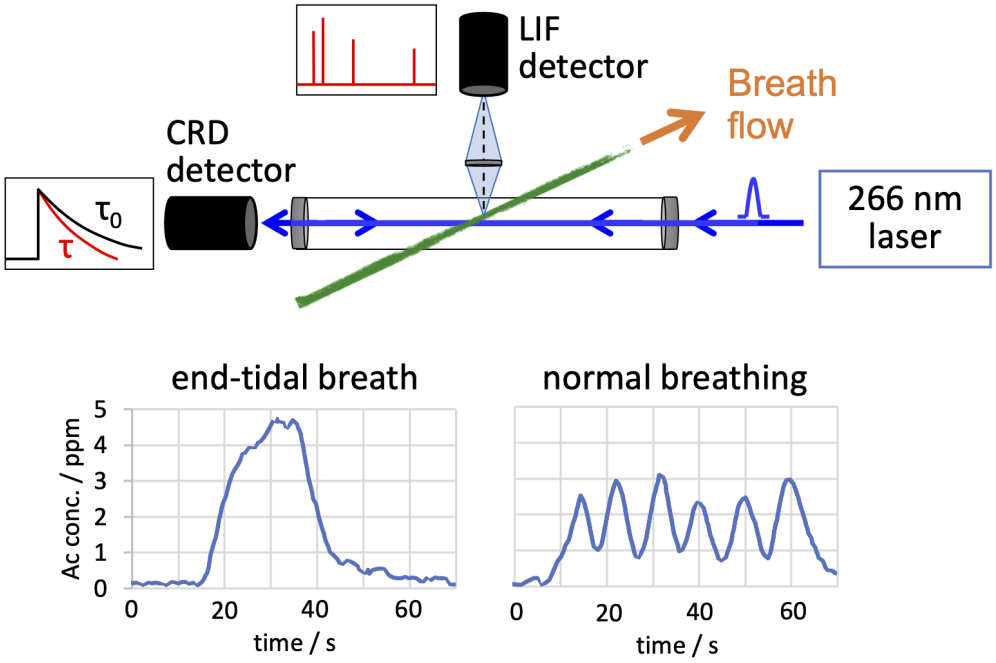
We have developed a cavity-enhanced laser-induced fluorescence (CELIF) sensor that monitors breath acetone in real time. The sensor allows to detect base-level acetone concentrations of healthy individuals (< 1 ppm) as well as extreme concentrations of > 1000 ppm found in severely ill patients with acute diabetic ketoacidosis (DKA). Accurate acetone concentrations are typically reported after a few breaths (approx. 30 seconds) from a passively breathing person.

Acetone is one of the trace volatile organic compounds found in human breath and is a biomarker



for raised blood ketone levels caused by increased fat-burning of the body's metabolism. Elevated breath-acetone levels are found in DKA patients or because of a low-carbohydrate (ketogenic) diet. The former constitutes a life-threatening condition that needs to be diagnosed and resolved in a timely manner. The latter is used to treat some medical conditions (e.g. epilepsy), for weight loss (fasting) or for conditioning for endurance sports.

The breath-acetone sensor is based on cavity-enhanced laser-induced fluorescence (CELIF), a spectroscopic technique that combines pulsed cavity ring-down spectroscopy (CRDS) with laser-induced fluorescence (LIF) detection in a single laser beam. The cavity provides the enhanced path length through the sample and acts as a very efficient spatial and optical filter for the fluorescence detection. In addition, the fluorescence signal can be calibrated directly via the reduced ring-down times at sufficiently high sample concentrations. Overall, CELIF provides superior limits of detection and dynamic ranges compared to a traditional CRDS, particularly for spatially confined samples [1, 2].

We will present first results obtained with our sensor from healthy individuals with base and

elevated breath-acetone levels to exemplify the performance and the potential of the sensor in clinical settings and for sports science.

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Real-world applications of Saturated-Absorption CAvity Ring-down (SCAR) spectroscopy

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Saturated-absorption cavity ring-down (SCAR) spectroscopy has pushed molecular detection to an unprecedented sensitivity of a few parts per quadrillion (ppq) [1], thus allowing precise quantification of an elusive isotopologue such as $^{14}CO_2$ (10^{-12} natural abundance). Technology has been progressing since its first demonstration in 2011 [2] and has taken to a portable instrument which is worldwide deployable. Recently, such a unique instrument has been applied for addressing specific problems in very different areas of science and humanities. Results will be shown for SCAR application to:

- discrimination of biogenic vs. fossil content in materials and fuels [3];
- radiological assessment of waste coming from decommissioning of a nuclear power plant [4];
- dating of archeological samples from a 4,500 years old Sumerian site.

Next applications aim to ^{14}C precise measurements in atmospheric samples. As is well known, CO_2 is the most significant anthropogenic greenhouse gas in the atmosphere. The pre-industrial level of 278 ppm represented a balance of fluxes among atmosphere, oceans and land biosphere. Currently, the global averaged CO_2 mole fraction has increased up to 413 ppm, mainly due to emissions from the combustion of fossil fuels and cement production. Therefore, distinguishing and measuring anthropogenic/fossil vs. biogenic CO_2 in the atmosphere is the key to quantify the anthropogenic contribution to climate change.

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The Miniature Airborne Cavity Enhanced Spectrometer (mACES) for sensitive NO_2 detection on UAV platforms

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Nitrogen dioxide (NO_2) is a criteria pollutant emitted from anthropogenic sources that is chemically converted in the atmosphere to O_3 and particulate matter, thereby affecting human health and the climate. NO_2 is highly reactive and has numerous sources, so strong gradients in its concentration can exist above the surface near populated areas. We present here a new instrument for measuring trace amounts of NO_2 on a UAV platform, which can access these populated areas with more versatility than traditional aircraft or tower platforms. The instrument, the Miniature Airborne Cavity Enhanced Spectrometer (mACES), measures absorption spectra from $425 - 475$ nm, which allows retrieval of NO_2 mixing ratios with a precision of less than 250 ppt in 1 second and with an accuracy better than 3%. This represents an improvement in performance over commonly used commercial electrochemical sensors. Additionally, mACES weighs less than 3 kg and draws less than 15 W power, and can be mounted on a hexacopter drone. We will present the results of the first tests of this instrument and discuss how it may be used in the future to make high-quality targeted measurements of NO_2.

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Applications of Cavity-based Polarimetry

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We describe how recent improvement in signal-reversing cavity-ringdown polarimetry [1, 2] can achieve optical-rotation sensitivities of order μdeg, and we give examples of applications including measuring chiral optical rotation from single drops, from thin films with effective thickness of about one monolayer, and from chiral emissions from a tree. We also discuss about the commercial potential of our compact instrument.

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Comb-assisted cavity ring-down spectroscopy for ultra-sensitive traceable measurements of water vapour in ultra-high purity gases

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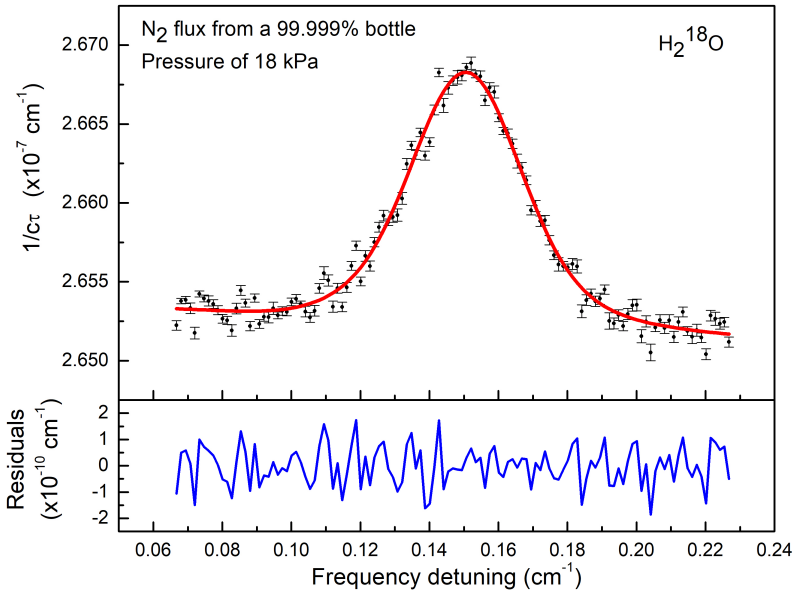
Water vapour is a critical contaminant for vacuum systems and ultra-high purity (UHP) gases used in many manufacturing processes such as the fabrication of semiconductor or optoelectronic devices. As water is a polar molecule, it is adsorbed on surfaces and then desorbed into any gas flow, thus affecting industrial processes significantly. Even at trace levels, it influences electrical, chemical and mechanical properties of materials, impacting on the quality and performance of semiconductor devices. Although it is considered the most difficult impurity to remove from gas distribution systems, the admitted levels of water in process gases such as N_2, O_2, H_2, He and Ar, are becoming increasingly low. In the framework of the European project PROMETH$_2$O (Metrology for trace water in ultra-pure process gases), we present here the first results of trace water measurements by means of a comb calibrated cavity ring-down spectrometer, operating at the wavelength of 1.39 μm. The spectrometer is based on a high-finesse ultra-stable optical resonator showing an empty-cavity ring-down time of about 130 μs. A first characterization of the set-up reveals a minimum detectable absorption coefficient of $9 \cdot 10^{-12}$ cm^{-1} and a noise equivalent absorption of $3 \cdot 10^{-11}$cm$^{-1}/\sqrt{Hz}$. We demonstrate the measurement of water vapour mole fractions at the level of 1 part per million (ppm) with a global uncertainty of 0.5%. The figure reported hereafter gives an example of $H_2{}^{18}O$



detection in a nitrogen flow, at a water mole fraction of about 1 ppm. The lower plot gives the residuals of a nonlinear least-squares fit to a Voigt profile. The signal-to-noise ratio was estimated to be about 20.

The spatial distribution of species in an atmospheric pressure plasma jet investigated by cw cavity ring-down spectroscopy

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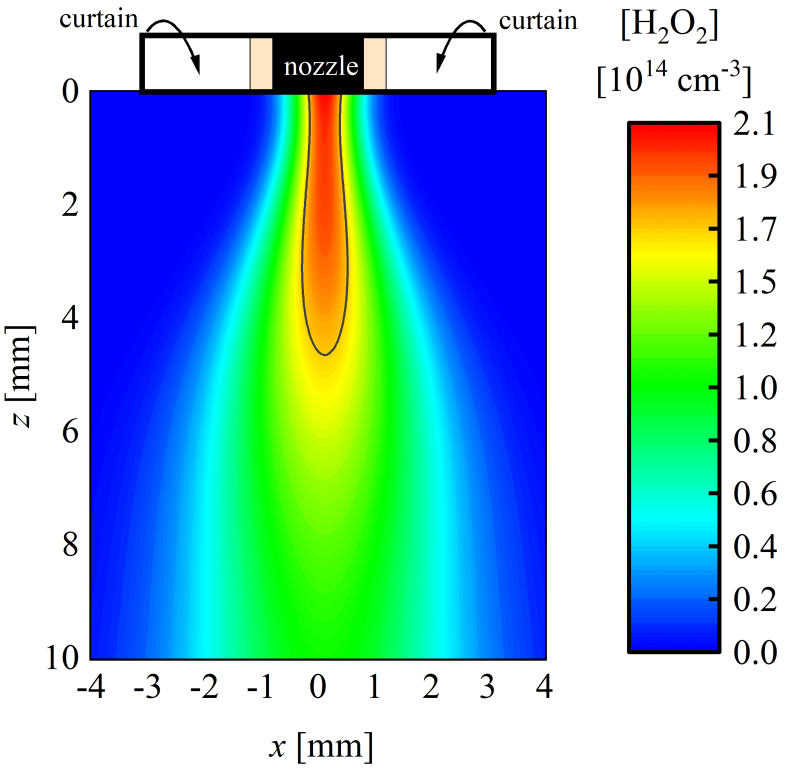
The application of cold atmospheric pressure plasma jets (CAPJs) for the localized treatment of heat-sensitive surfaces and biological tissue has gained high industrial and medical importance. A detailed understanding of the complex chemical reaction network confined to a CAPJ would enable customized compositions of reactive species to be tailored for a specific plasma application. This requires highly sensitive and selective measurements of transient molecules and free radicals, their spatial and temporal distributions, and their transport in non-equilibrium environments. However, some of the key radical species are often present only in trace amounts and for a short period of time, although they often drive the chemistry and therefore their detection is crucial. A challenge for diagnostics of CAPJs is the small dimensions (µm to cm range) over which the reactive species are confined. A common approach to obtain absolute species densities are absorption



spectroscopy techniques. To increase the absorption path length, cavity-enhanced spectroscopy methods can be applied. However, with these techniques often line-of-sight densities without any spatial information are obtained. Nevertheless, using continuous-wave cavity ring-down spectroscopy, we were able to determine the spatial distribution of the HO_2 radical [1, 2] and the H_2O_2 molecule in the effluent of the plasma jet CAPJ device kINPen (see figure), developed at the INP for use in plasma medicine.

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Ultra-high sensitivity and ultra-wide dynamic range gas detection based on photoacoustic spectroscopy

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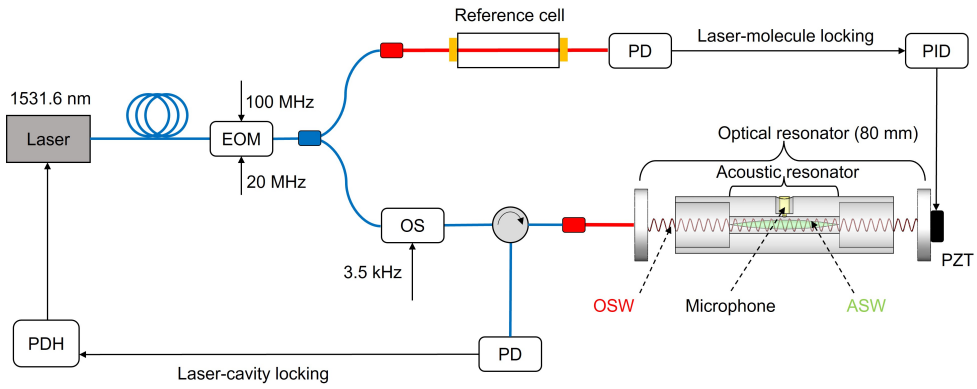
Photoacoustic spectroscopy (PAS) is a promising method for gas sensors with high sensitivity, fast response, low cost, and a small footprint. However, compared to the well-assessed cavity ring-down spectroscopy and noise-immune cavity-enhanced optical-heterodyne molecular spectroscopy techniques ($10^{-14} - 10^{-12}$ cm^{-1} in noise equivalent absorption (NEA)), the NEA of state-of-the-art PAS gas sensors is limited to $10^{-11} - 10^{-8}$ cm^{-1}, while the dynamic range is limited to five orders of magnitude [1–6]. Herein, we report a method to simultaneously enhance the acoustic and optical waves using combined optical and acoustic resonators in a centimeter-long configuration. The photoacoustic signal can be enhanced linearly by orders of magnitude via double standing waves inside the resonators. Not only the lower detection limit is enhanced by the double standing waves, but also the upper detection limit is expanded due to the short resonators. As shown in figure, we developed a sensor by detecting acetylene (C_2H_2), achieving a noise equivalent concentration (NEC) of 1.75 ppt, an NEA of $1.9 \cdot 10^{-12}$ cm^{-1} and a dynamic range of seven orders of magnitude.



Schematic representation of the experimental apparatus. EOM: electro-optic modulator; OS: optical switch; OSW: optical standing wave; ASW: acoustic standing wave.

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Molecules in optical cavities: precision spectroscopy and strong light-matter interactions

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Direct frequency comb spectroscopy is a sensitive, broadband, and precision technique that can be used to interrogate the quantum structure of unprecedentedly large molecular species. Cavity-enhanced frequency comb spectroscopy (CE-FCS) matches the evenly spaced spectral structure of a frequency comb light source to the resonant modes of a high-finesse optical cavity. This method allows for simultaneous detection of absorption signal across the comb spectrum, extremely high frequency resolution, and high sensitivity through cavity-enhancement. We combined CE-FCS with cryogenic buffer gas cooling to measure the rovibrational structure of buckminsterfullerene (C_{60}). These measurements represent the first probe of the quantum structure of C_{60} and establish it as by far the largest molecule for which a state-resolved spectrum has been reported [1]. We subsequently implemented a cw mid-IR spectrometer based on a quantum cascade laser locked to a high-finesse cavity using optical feedback stabilization. Using this apparatus, we report new nonlinear saturation spectra of C_{60} which illuminate state-specific vibrational and rotational energy transfer and relaxation processes as molecules undergo collisions with a range of buffer gas collision partners.

For applications in cavity-enhanced spectroscopy, optical cavities enhance the strength of light-matter interactions and thereby increase the sensitivity of absorption measurements. Some of the Weichman Lab's research interests lie in a more exotic regime for intracavity molecules. The interaction of light and matter is typically weak and can be treated perturbatively. This picture breaks down in the regime of strong coupling, where the rate of light-matter interaction competes with the dissipation of excitations. In this regime, superposition states with mixed light-matter character, dubbed polaritons, emerge. Polaritons inherit the coherent, wavelike nature of light while maintaining local molecular interactions and structure. Polaritonic molecules may therefore demonstrate distinct reactivity from their ordinary uncoupled counterparts, representing a rich sandbox for chemistry. In our new lab at Princeton, we are setting out to understand how molecular polaritons behave and react, using ultrafast and precision spectroscopy to follow the reaction dynamics of benchmark condensed-phase and gas-phase systems under strong light-matter coupling.

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Mode-resolved Mid-infrared optical frequency comb spectroscopy using an air-spaced VIPA without optical cavity filtering

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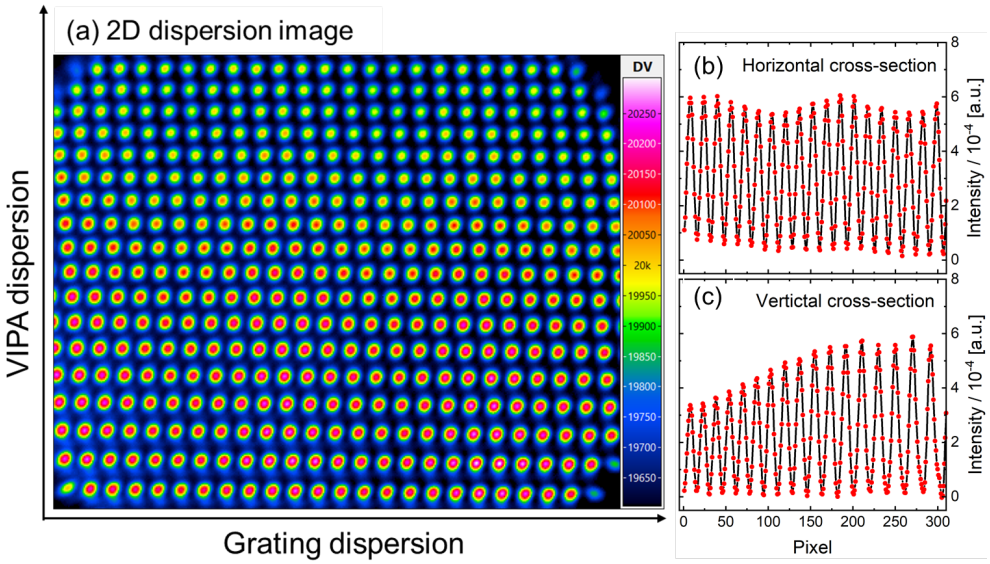
The cross-dispersion of a virtually imaged phased array (VIPA) etalon together with a grating, which is commonly known as a VIPA spectrometer, drastically improves the spectral resolution of conventional grating-based spectrometers. VIPA spectrometers based on optical frequency combs have proven versatile for high spectral resolution and chemical kinetics measurements [1, 2]. Previous demonstrations for resolving the modes of a frequency comb using VIPA spectrometers involved the use of an optical cavity to filter them, hence increasing their spacing relative to the VIPA resolution. Such an approach could be proven to be useful for high spectral accuracy measurements. However, the increased time needed to measure spectra at several repetition rates, f_{rep}, limits the applicability of this approach to perform time-resolved measurements under low pressure conditions, where Doppler-limited absorption profiles are the target of interest.

In this work, we report on the first air-spaced VIPA spectrometer using a mid-infrared frequency



(a) 2D dispersion image of resolved comb modes with a 250 MHz f_{rep} measured by the air-spaced VIPA spectrometer. (b,c) horizontal and vertical cross-sections of the 2D image.

comb. The new spectrometer is capable of fully resolving the modes of a frequency comb with a $f_{rep} = 250$ MHz without the use of a filtering optical cavity, see figure. The use of an air-spaced etalon was essential for achieving such a high resolution as intrinsic temperature-driven etalon effects of previously used solid VIPA etalons limit their effective resolution, in particular at unstabilized temperature conditions. The spectrometer is tested by measuring Doppler-limited absorption profiles of CH_4, around 3000cm^{-1} and HCN, around 3250 cm^{-1}, generated in a plasma nitrocarburizing process at a pressure of 1.5 mbar and gas flows of 20 sccm H_2, 20 sccm N_2, and 2 sccm of CH_4. Future work will be focused on introducing an optical cavity to measure key transient species in the plasma, hence elucidate the complex plasma-induced chemistry.

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High-accuracy line positions of N_2O, CH_4 and H_2CO around 8 μm from optical frequency comb Fourier transform spectroscopy

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The atmospheric window at $8 - 12$ μm contains vibrational absorption bands of many small molecules while being largely free from interfering water absorption. Apart from atmospheric sensing, this spectral range is also important for astrophysics and falls within the detection range of the recently launched James Webb Space Telescope. However, spectroscopic databases at these wavelengths are based mostly on traditional FTIR measurements, with line position uncertainties of the order of a few MHz. Significant improvements in precision and accuracy can be achieved using optical frequency comb spectroscopy that provides a combination of accurate frequency scale and broad optical bandwidth. Here, we present high-resolution absorption measurements of N_2O, CH_4 and H_2CO between 1240 and 1380 cm^{-1} obtained with a recently developed Fourier transform spectrometer [1] based on an 8 μm difference frequency generation comb source [2] and a multipass cell. Using the sub-nominal sampling-interleaving scheme [3] and fitting the spectra line by line allows determination of line center frequencies with uncertainties of a few hundred kHz. For the ν_1 band of N_2O [1], we observed excellent agreement in the retrieved line positions with a previous high-accuracy study [4]. For methane, a major greenhouse gas and a constituent of exoplanetary atmospheres, we determined center frequencies of over 800 lines belonging to the ν_4; fundamental band of $^{12}CH_4$ and $^{13}CH_4$ as well as two $^{12}CH_4$ hot bands, with 30 times better precision than the previously available data. Including these results in a global fit of the methane spectrum yielded a strong reduction in residuals. Currently, we measure the spectra of the ν_4 and ν_6 bands of formaldehyde, H_2CO, a toxic pollutant also found in the interstellar medium, for which publicly available data have been lacking in this spectral region.

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Thursday, June 16th

Understanding ultrafast observables via broadband cavity-enhanced ultrafast spectroscopy in jet-cooled molecules and clusters

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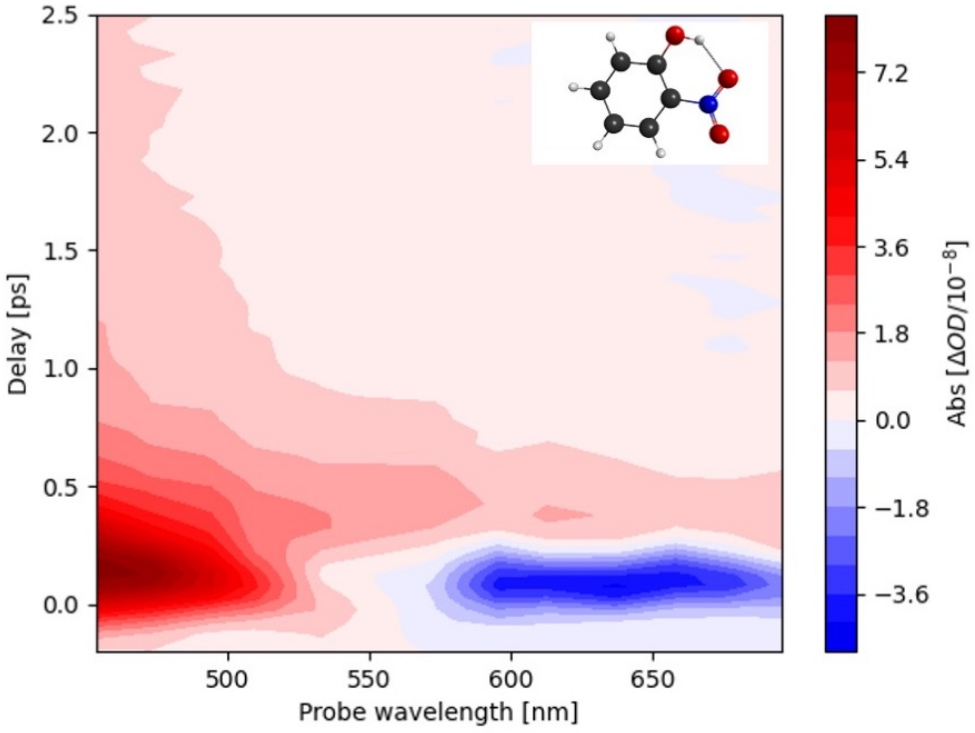
Ultrafast spectroscopy offers the opportunity to directly probe the dynamics of molecules after excitation. However, the interpretation of ultrafast experiments remains a challenge because of the projection of highly dimensional dynamics into a much lower dimensional signal is unavoidable. Using frequency comb lasers and optical resonators, we have developed a broadband ultrafast transient absorption spectrometer with high sensitivity, enabling all-optical measurements in molecular beams. This enables direct comparisons of data recorded from jet-cooled molecules and clusters with molecules in solution. By performing studies on a series of archetypal molecules in collaboration with others working on molecular dynamics theory and time-resolved photoelectron spectroscopy, we aim to get a clearer picture of the meaning and correct interpretation of ultrafast observables in general.

I will present ultrafast spectroscopy results on several molecules undergoing excited-state proton



transfer, internal conversion, and intersystem crossing, and the effect of Ar clustering on these dynamics. I will also discuss several technical details of the broadband cavity-enhanced spectrometer, many of which can be useful for those performing cavity-enhanced spectroscopy with high frequency resolution as well.

If there is time, I will also discuss recent theoretical work regarding rotationally-resolved 2DIR spectroscopy.

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High finesse Fourier transform cavity ring-down spectroscopy with an optical frequency comb

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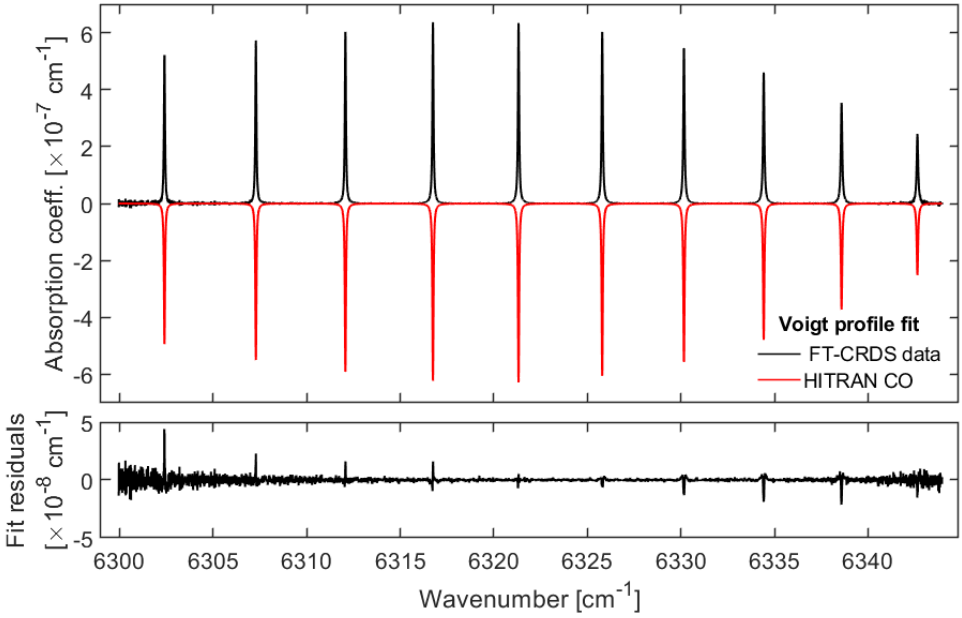
Cavity ring-down spectroscopy (CRDS) is a very sensitive and widespread spectroscopic technique, mainly used with continuous-wave lasers. Multiplexed CRDS could offer broadband and calibration-free spectra but requires a spectrometer able to separate the exponential decays of each spectral element [1, 2]. We recently demonstrated CRDS spectrometer based on an optical frequency comb and a time-resolved Fourier transform spectrometer (FT-CRDS) [3]. The Er:fiber comb was locked to a $2,000$ finesse cavity using the Pound-Drever-Hall technique to ensure a quasi-continuous transmission. This setup was used to detect atmospheric CO_2 and H_2O. We will present the ideas behind the technique, apparatus, and data processing. However, in that demonstration the cavity length was left free-running, preventing any averaging. Moreover, switching off the light to trigger ring-down events also interrupted the comb-cavity locking, limiting the usable cavity finesse. We



will introduce a new FT-CRDS setup allowing for cavity finesse of $20,000$. In this approach, the comb and cavity are referenced to a cw laser to ensure absolute stabilization of both while isolating the error signal from the CRDS decays. We performed spectroscopy of 250 ppm of CO diluted in Ar at 534 Torr. We averaged 15 spectra recorded with a spectral resolution of 236 MHz, each acquired in 5 min 16 s. The ring-down spectrum is shown in the figure together with a model based on Voigt absorption profiles and HITRAN parameters. The noise equivalent absorption is equal to $7.0 \cdot 10^{-10}$ cm^{-1}, and the spectrum contains 4400 resolved elements, yielding a figure of merit of $7.3 \cdot 10^{-10}$ cm^{-1}Hz$^{-\frac{1}{2}}$ per spectral element. The sensitivity is high enough to observe speed-dependent effects, visible in the residuum of the fit to the measured spectrum. We will discuss the performance improvement and the spectroscopic analysis.

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Electro-optical dual-comb cavity ring-down, mode-width and mode-dispersion spectroscopy

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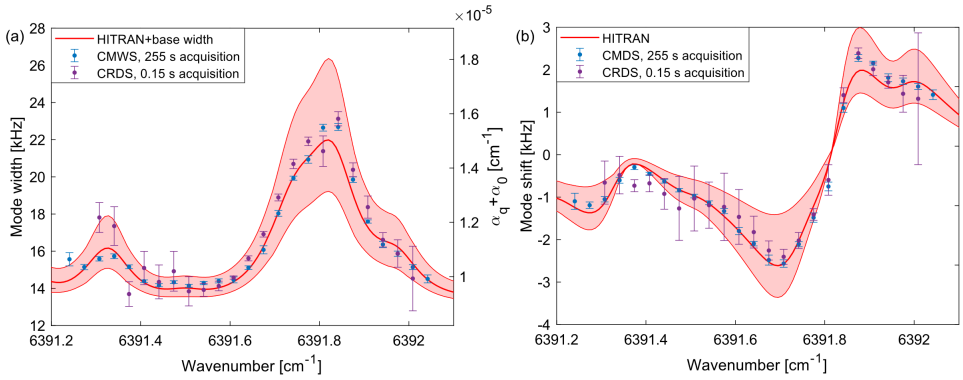
Cavity-enhanced spectroscopic techniques based on frequency combs are rapidly developing, bringing the possibility of broadband and parallel measurements to methods first shown with continuous wave (cw) lasers: CRDS [1, 2], CMWS and CMDS [3, 4]. We present the first demonstrations [5, 6] of these techniques combined with dual-comb spectroscopy. We propose a new approach to CRDS, in which time constants of cavity ring-downs are determined not directly from the intensity decays, but from Lorentzian shapes of the cavity modes visible in the Fourier-domain spectrum down-converted from optical to radio frequencies. All the measurements were performed with



a versatile experimental setup based on a single cw laser, used both to generate two frequency combs with electro- and acousto-optical modulation and to stabilize the frequencies of the combs. Two electrical switches were used to turn on and off the light of the combs to trigger the acquisition of ring-down signals, while changing the AOM frequencies allows for tuning the combs over the cavity modes to perform the CMWS/CMDS measurements.
Parameters of the cavity modes are used to determine absorption and dispersion spectra. Contrary to conventional CRDS, this scheme allows retrieval of dispersive information. The figure presents spectra of 760 Torr of a mixture of 20% methane in nitrogen. (a) shows an absorption spectrum obtained with CMWS and CRDS, while (b) presents the dispersion spectrum obtained with CMDS and CRDS techniques. All spectra remain in good agreement with the simulation based on HITRAN (red regions represent calculated error margins) [7].

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Cavity-enhanced double-resonance spectroscopy of methane using a frequency comb probe

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Spectra of various astrophysical objects contain signatures of hot molecules. Theoretical models of high-temperature molecular spectra are necessary to interpret these observations [1, 2]. These models, in turn, need to be validated by accurate laboratory measurements and assignments of hot-band transitions. We developed optical-optical double-resonance spectroscopy with a frequency comb probe to measure previously unobserved sub-Doppler hot-band transitions in the $3\nu_3 \longleftarrow \nu_3$ region of methane with line position accuracy of ~ 1.7 MHz [3, 4]. The methane sample was contained in a liquid-nitrogen cooled single-pass cell. The pump was the idler of a singly-resonant continuous-wave optical parametric oscillator stabilized to a Lamb dip in a selected CH_4 transition in the ν_3 band ($\sim 3.3\,\mu$m), and the probe was an Er:fiber frequency comb, shifted to $1.67\,\mu$m by a polarization maintaining microstructured silica fiber. The comb spectrum was detected by a Fourier transform spectrometer with auto-balanced detection. Using the sub-nominal sampling-interleaving scheme, we measured probe spectra covering 6 THz with sampling point spacing of 2 MHz. The measured probe transition frequencies served as the first verification of theoretical predictions in this range, and agreed within 30 GHz with the data from the TheoReTS database [1]. To increase the absorption sensitivity and line position accuracy we recently implemented a room-temperature enhancement cavity for the comb probe. Compared to the initial demonstration, the absorption sensitivity is now increased by more than two orders of magnitude, which allows detection of weaker transitions with better signal-to-noise ratios, and the accuracy of the line positions is improved to 1 MHz. We assign the probe transitions by comparing line intensity ratios measured with parallel and perpendicular relative pump/probe polarizations, and by reaching the same final state with different pump/probe configurations. The cavity-enhanced system allows the detection and assignment of new hot-band transitions, and investigation of sub-Doppler probe transition line shapes.

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Microbubble resonators as enhanced platforms for thermometry and flow cytometry

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CNR-IFAC, Institute of Applied Physics, Carrara, Italy

Over recent years, photoacoustics has drawn attention for innovative applications in biomedical optics. Most photoacoustic platforms rely on piezo transducers that were developed within the venerable context of ultrasonography. However, the generation or propagation of acoustic signals in photoacoustics and ultrasonics differ in fundamental aspects that deserve innovative solutions and yield new opportunities. Here, we disclose our design and results for an all-optical photoacoustic flow spectro-cytometer that may be exploited in multiple contexts, such as the detection of circulating cells in bio fluids as well as the inspection of colloidal suspensions of artificial particles in sols or aerosols. This setup rests on the implementation of an optical microcavity resonator that may be coupled to a microfluidic interface and filled with any sample of interest. Then, its excitation with short optical pulses triggers a photoacoustic event within the cavity, which imparts a transient deformation of the glass resonator and its dielectric landscape, thus shifting its optical resonances. The advantages of this setup include its inherent feasibility for miniaturization and workability in air rather than water, as occurs with piezo transducers for ultrasonography. We illustrate these features in the analysis of the spectral fingerprints of colloidal suspensions of plasmonic particles with volumes in the nL range and repetition rates in the kHz domain [1]. We also present a thermal detection that produces spectra insensitive towards light scattering in the sample, as proved experimentally by comparing the spectra of acqueos gold nanorods suspensions in presence or absence of milk powder [2]. In perspective, this system could be implemented for the characterization of turbid biological fluids through their optical absorption, especially when considering that the microbubble resonator naturally interfaces to a microfluidic circuit, and may easily fit within portable or on-chip devices.

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Optomechanical devices for high accuracy acceleration sensing

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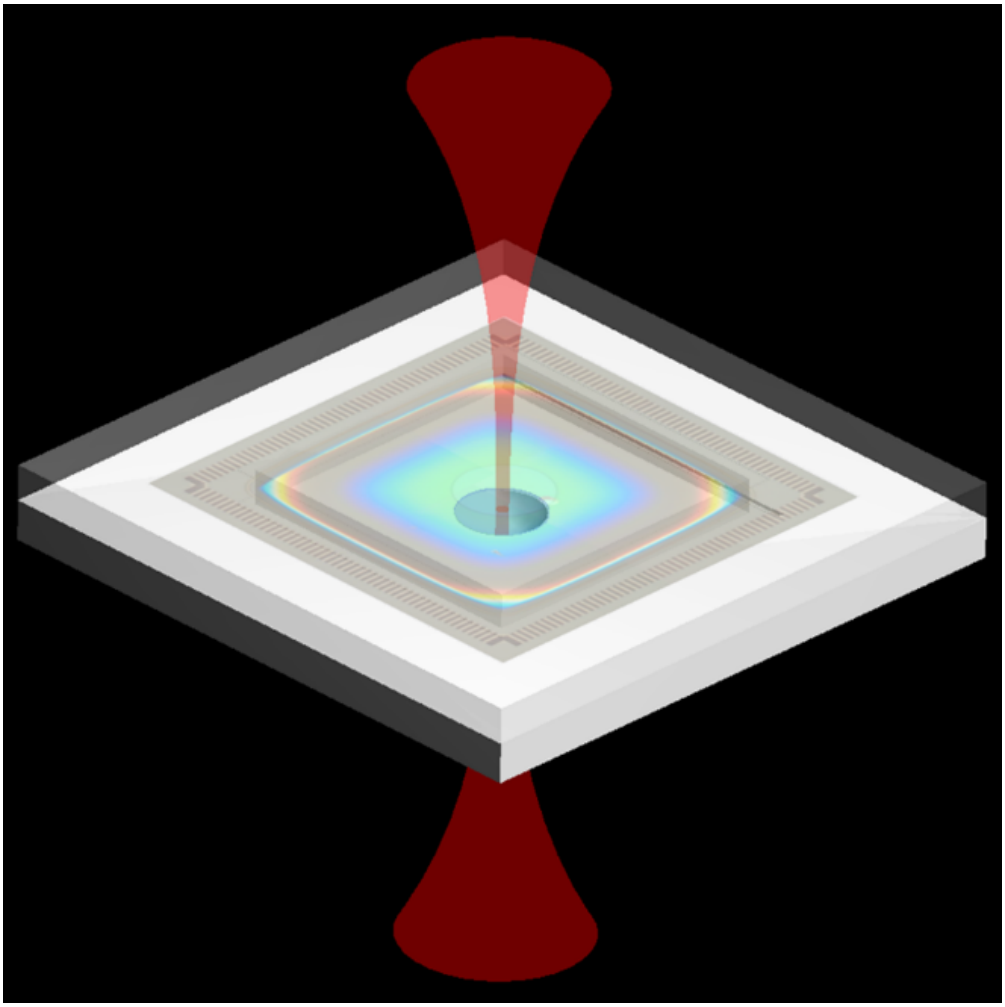
Optomechanical cavities offer exceptional sensitivity to external stimuli such as temperature and pressure changes as well as acceleration and acoustics. Recently, we have developed Fabry-Perot optical microcavities in which one of the mirrors is a proof mass suspended by microbeams [1, 2]. These devices exhibit acceleration sensitivity as high as 32 ng/Hz$^{1/2}$ limited only by thermomechanical noise. In addition, due to the presence of only a single low frequency vibrational mode, the measured displacement can readily be converted into acceleration, paving the way for use as an intrinsic standard. Further, I will discuss novel readout approaches which leverage electro-optic frequency combs for rapid, high dynamic range interrogation [3, 4].



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High-finesse optical cavity length adjustment at cryogenic temperatures

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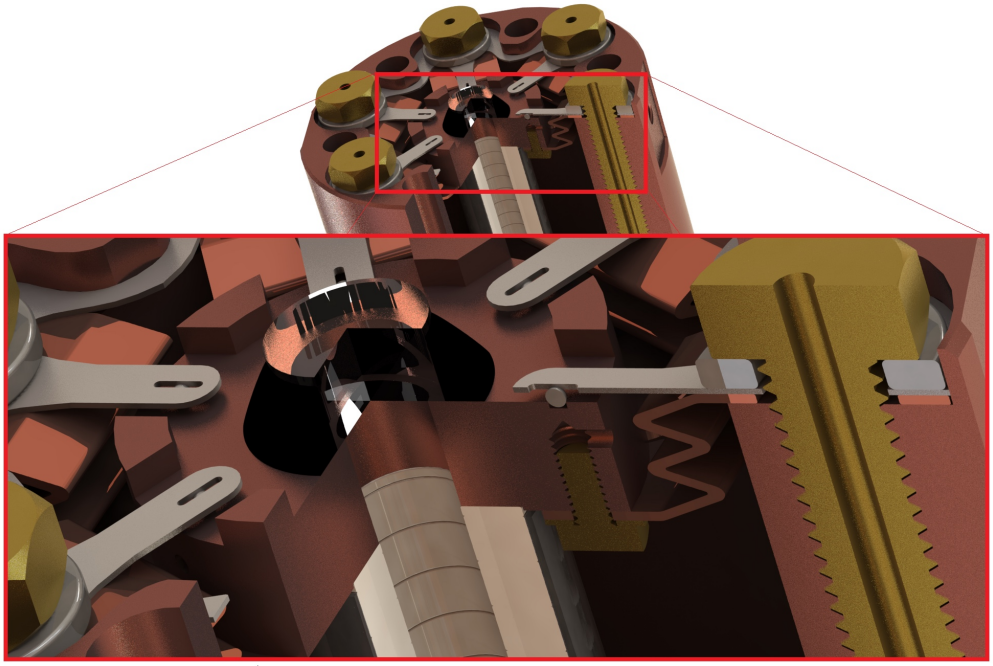
High-finesse optical cavities offer an obvious profit for molecular spectroscopy, which is ultra-long path of molecule-light interaction that directly determines high sensitivity of the technique. A crucial component that makes high-finesse cavities suitable for molecular spectroscopy applications is an actuator that controls the length of the cavity to compensate for cavity length drifts and enables probing the spectrum at grid denser than free spectral range [1, 2]. Spectrometers working at low temperatures allow precise determination of spectra for large molecules and achieve high accuracy of measurements conducted for simple molecules, which is important in fundamental studies. When designing experiments of this type a major challenge is to select optomechanical components that can operate under required range of temperatures. We demonstrate a method for controlling the length of an optical cavity in a wide temperature range, from room temperature to cryogenic regime (10 K). We conduct tests to verify proposed solution and to determine its effectiveness over the entire temperature range.



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Cavity-enhanced vs coherent Raman metrology of H_2

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Molecular hydrogen is a benchmark system to test quantum electrodynamics and the standard model of particle physics at molecular length scales. Thanks to its simplicity, calculations of rovibrational energy levels can be carried out with part-per-billion uncertainties. However, determination of transition frequencies through absorption spectroscopy is a difficult task due to the lack of a molecular dipole moment. Moreover, line shapes are atypical and difficult to model due to very strong collisional effects contributing strong asymmetry that prevents straightforward extrapolation of the transition frequency of the unperturbed molecule. To overcome such problems, one needs to employ high sensitivity spectrometers, and to use advanced modeling to reduce the impact of collisional effects on the determination of transition frequencies. In our work we adopted two different strategies to reach high signal to noise ratio even at relatively low pressures, exploiting a highly sensitive cavity ring-down spectrometer to measure overtone transitions in the near infrared and developing a novel comb-referenced stimulated Raman spectrometer to probe fundamental transitions in the mid infrared. In the first case, we investigated the $_2{-}_0$ S(3) and S(4) transitions of D_2, lying around $1.57\ \mu$m spectra were measured in a wide pressure range spanning 0.13 to 1.2 bar using a cavity ring-down spectrometer linked to an optical frequency comb. We modeled the complex lineshape through the so-called β-corrected Hartmann-Tran profile, performing a multi-pressure fit of the measured absorption lines to reduce correlations between collisional parameters involved in the fit. Moreover, we reduced the number of fit parameters by fixing some of them to values obtained from *ab-initio* quantum scattering calculations. The determined transition frequencies have an uncertainty of 900 kHz, a 30-fold improvement with respect to the experimental state of the art. In the second case, we performed nonlinear spectroscopy through stimulated Raman scattering exploiting two narrow-linewidth near-infrared lasers as the pump and Stokes sources to probe the $_1{-}_0$ Q(1) transition of H_2 around $2.4\ \mu$m. The approach uses an optical frequency comb to calibrate the frequency spacing between the pump and Stokes beams that interact with H_2 in a multipass cell. We performed measurements in a pressure range spanning 0.05 to 4 bar, which fully covers the transition from the low-pressure, mostly Gaussian lineshape to the Dicke-narrowed Lorentzian lineshape occurring around 2.5 bar. We exploited such a wealth of information to develop a robust extrapolation procedure for the unperturbed transition frequency, reaching an uncertainty of 300 kHz, a 20-fold improvement with respect to the state of the art and a factor of 2 better than the most recent theoretical determination.

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Infrared off-axis cavity-enhanced absorption spectroscopy of molecular hydrogen

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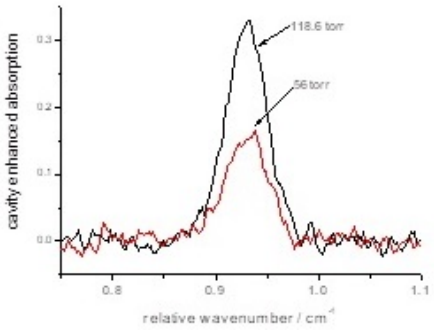
Infrared laser spectroscopy is often used for measuring low concentrations of trace gases but hydrogen does not exhibit electric dipole moment transitions in the infrared spectral region. However, it does possess a weak quadrupole moment which can be used if long optical absorption pathlengths are employed. This work describes an off-axis cavity-enhanced absorption experiment to measure hydrogen around 2.1μm. Quadrupole transitions have the rotational selection rule J$= 0, \pm 2$ which gives rise to a Q branch as well as S and O branches. The quadrupole moment spectrum of H_2 was first measured by Herzberg in 1949 [1] and later by Fink *et al.* [2] The S(1) line of the fundamental vibrational quadrupole band has a linestrength of $3.36 \cdot 10^{-26}$ cm/molecule at 298 K.
A cw-DFB diode laser producing around 5 mW at 2122 nm was used to illuminate a high finesse optical cavity of length 19.4 cm in an off-axis alignment. The laser light emerging from the cavity was focused onto an extended InGaAs photodetector. Difficulties were encountered due to amplified spontaneous emission of the laser passing through the cavity onto the detector. Hydrogen gas (99.99%) was introduced into the optical cavity using a gas sample bag and the infrared spectrum at several different pressures was measured. Figure shows an example of the spectra obtained



with the off-axis cavity enhanced absorption technique. The very low laser power detected at exit of the cavity limited the detection limit to 0.16% hydrogen for 200 scan averages within 2 s. This was limited by detector noise. Higher laser power and a longer effective pathlength should enable detection limits lower than ppm levels.
In conclusion, we have demonstrated that off-axis cavity enhanced absorption spectroscopy can be used to quantitatively measure molecular hydrogen in the infrared spectral range. The technique can now be incorporated into the atmospheric pressure plasma catalysis reactor to study the formation of hydrogen.

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Fast breath analysis by OF-CEAS: clinical studies for lung transplantation

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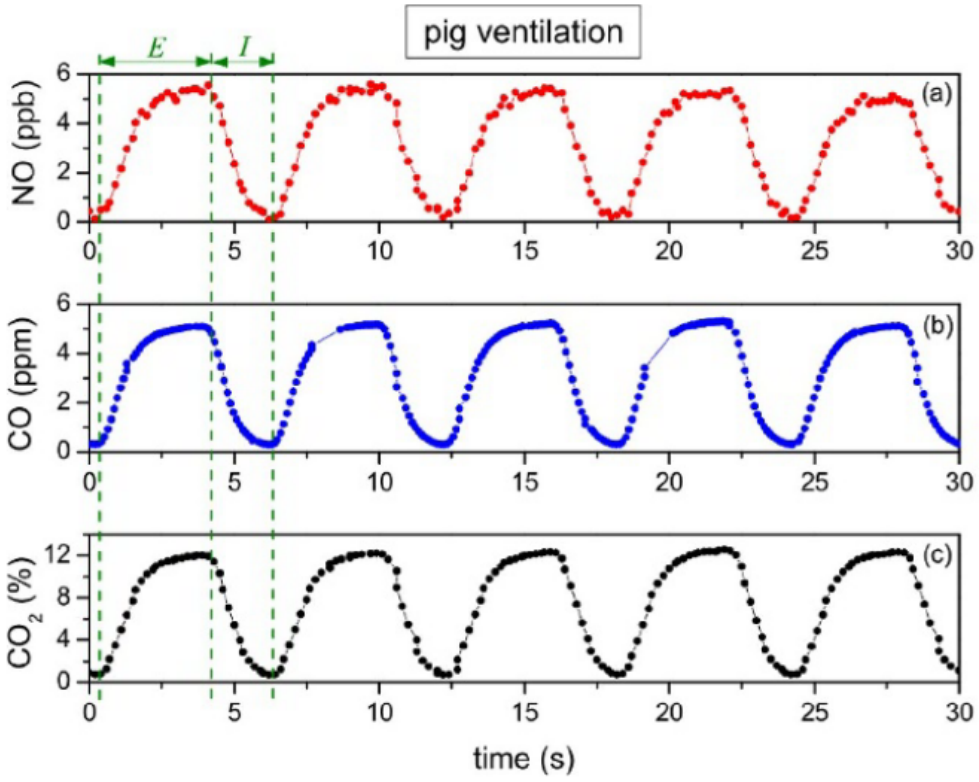
The sense of smell is one of the oldest diagnostic tools in the practice of medicine. In the last decades, exhaled air analysis for non-invasive medical diagnosis has seen a growing interest. There are a thousand species present at very low concentrations in exhaled air, of which around thirty have been identified as markers of physiological processes and of various pathologies [1]. Among the different analytical methods, tunable diode laser spectroscopy appears to be very attractive. It allows fast response, low detection limits and excellent selectivity. Numerous laboratory demonstrations have been carried out with different techniques based on absorption spectroscopy in multi-pass cells or with resonant cavities [2, 3]. Today, we are at a turning point where some state-of-the-art analyzers developed in the laboratory are finding well-identified medical applications, thanks to close collaborations between physicists and doctors engaged in clinical studies. Optical-Feedback Cavity-Enhanced Absorption Spectroscopy (OF-CEAS) is a technique introduced



(a) Nitric oxide, (b) carbon monoxide and (c) carbon dioxide measured in real time by OF-CEAS when a pig is ventilated. Regular respiratory cycles of 6 s duration are recorded with an inspiration (I) to an expiration E ratio: $I/E = 1/2$

in our group [4], allowing realization of robust and compact gas analyzers, exploited in several

interdisciplinary applications and collaborations.
Adaptation of OF-CEAS commercial instruments (by AP2E) allowed to perform pre-clinical (animal models) [5] and, more recently, clinical studies (on human patients) [6] in the context of lung transplantation. The objective is to investigate new markers to assess the quality of lung grafts for their requalification in order to increase the number of available grafts. Real-time measurements (with single respiratory cycle time-resolution) of the production of CO, NO and CO_2 by ex-vivo lungs are carried out by OF-CEAS (see figure).

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Friday, June 17th

Spectroscopy and control of individual erbium dopants in optical resonators

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In spite of decade-long research into different physical systems, the demonstration of a scalable platform for quantum networks and distributed quantum information processing remains an outstanding challenge. In this context, our group investigates the use of erbium dopants in silicon [1, 2] and silicate crystals [3–6]. As the optical transition of erbium is the narrowest spectral feature ever measured in a solid, spectroscopy requires a stable laser, which we implement via a cryogenic fiber ring resonator [7]. In addition, the dopants need to be embedded into a cryogenic resonator with small mode volume and large quality factor. Using a Fabry-Perot cavity with a finesse of 100000 we could demonstrate up to 70-fold Purcell enhancement with lifetime-limited optical coherence in the telecommunications frequency window [3]. In recent experiments, we could observe and coherently control about 100 individual dopants with ultra-low spectral diffusion, ~ 0.1 MHz, determined by the nuclear spin bath [6]. Using nuclear spin initialization or silicon instead of silicate host crystals may further reduce this value below the 20 kHz homogeneous linewidth we observed recently in a nanophotonic waveguide [2]. This would enable the realization of entanglement between remote dopants with high efficiency and fidelity. Thus, our novel hardware platform may facilitate the implementation of scalable quantum networks and repeaters based on single emitters at telecom wavelength.

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Broadband nonlinear cavity ringdown spectroscopy

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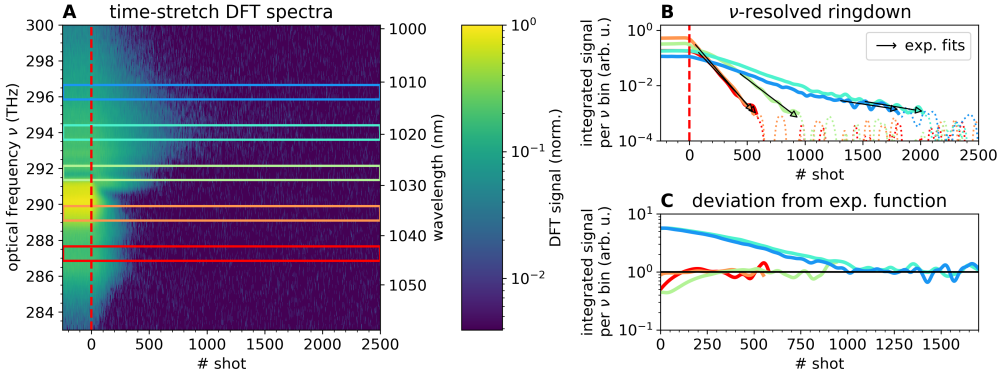
In traditional cavity ringdown spectroscopy (CRDS), a passive optical resonator is first loaded with light. Upon rapidly turning off the input light, the exponential intensity decay over hundreds of roundtrips is then measured with very high sensitivity and precision, unaffected by laser noise. Consequently, CRDS has been widely used for applications such as trace-gas detection or loss characterization of highly-reflective laser mirrors.
Here, we report nonlinear CRDS of pulse-train-driven, free-space cavity solitons [1] and use a dispersive-Fourier-transform (DFT) single-shot spectrometer [2] to record each individual pulse of the decaying soliton. The (non-exponential) per-wavelength decay curves contain rich information about the optical nonlinearities governing the formation of the self-stabilizing, self-compressing soliton state. This is, to the best of our knowledge, the first application of CRDS to nonlinearities driven by ultrashort pulses.
In our experiment, a 100-W cavity soliton is excited in a 73-MHz femtosecond enhancement cavity



(A) Nonlinear CRDS signal. (B) Optical-frequency-integrated data. (C) Relative deviation from the exponential functions obtained from the low-intensity part of the measured decay.

(fsEC), in analogy to [1]. A low-power (20 mW) copy of the circulating beam is sent to the single-shot spectrometer, comprising 2.5 km of glass fibre, and a fast photodiode and oscilloscope. The fibre's dispersion maps the optical frequencies of the incoming pulses to the (measured) arrival times at its end. After abruptly interrupting the seed, the spectrum of each individual roundtrip of the decaying soliton can thus be measured (figure a). The deviation from the exponential behaviour typical for linear CRDS (figure b,c) entails the nonlinear response of the sapphire plate in the fsEC focus, probed at a series of diminishing intensities. Our approach allows high-precision measurements of the instantaneous (Kerr) and non-instantaneous (Raman) 3^{rd}-order nonlinearities. We expect this to benefit the design of nonlinear resonators supporting shorter solitons, and, potentially, of other nonlinearities, e.g., plasma formation in cavity-enhanced high-harmonic generation [3].

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Microcavity-enhanced investigation of nanoparticle dynamics

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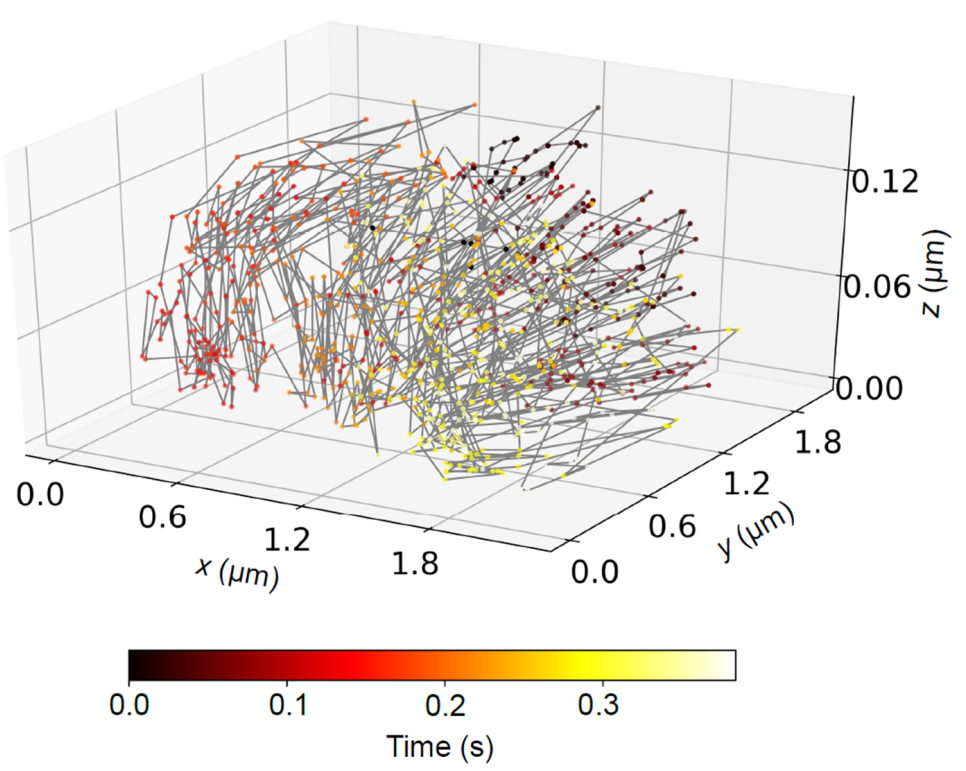
We have developed a fibre-based high-finesse Fabry-Perot microcavity capable of detecting and characterising single nanoparticles. While many current nano-detection techniques depend on labelling, our sensor can detect unlabelled nanoparticles in dispersion. Silica nanospheres with radii of 25 nm and gold nanorods 20 nm in length have thus been detected in water.
Properties of the nanospheres such as the size, polarizability and refractive index were deduced, and the three-dimensional Brownian motion of a single nanoparticle in the cavity was tracked by simultaneous measurement of the fundamental and higher-order transverse resonant modes. The best spatial and temporal resolutions of the particle's motion achieved were 8 nm and 0.3 ms respectively. Furthermore, the rotation of nanoparticles could be measured using the polarisation splitting of the fundamental mode.[1–3] An active cavity stabilisation scheme has been



3D track of a nanosphere undergoing Brownian diffusion

implemented which can achieve an rms jitter of 4% of the linewidth in a water-filled microcavity having a finesse of over $5 \cdot 10^4$. When cavity scanning is too slow to resolve small dynamic time constants, e.g. the rotation of gold nanorods, measurement using a locked cavity can be used to provide a high measurement bandwidth limited only by the detector or the cavity linewidth. This enables the investigation of fast nanoparticle dynamics and additionally offers the advantages of improved mechanical stability and drift compensation.
We shall report progress towards the quantitative measurement of nanorod dynamics, and imple-

menting cavity-locked high-speed polarisation-splitting measurement techniques. Based on this, we aim to investigate the optical and dynamic behaviour of single biomolecules, such as DNA.

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Single-molecule thermo-optoplasmonic sensing of enzymes

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Optical sensors based on whispering-gallery-mode (WGM) cavities are exceptional tools for detecting molecules down to single levels. WGM cavities coupled to plasmonic nanorods, or optoplasmonic sensors, have proven their ability to detect single molecules mostly via reactive sensing when WGM frequency is tracked. In this contribution, we discuss several mechanisms for single-molecule detection with optoplasmonic sensors including newly developed thermo-optoplasmonic sensing. As objects of investigation, 3-phosphoglycerate kinase (3PGK) was chosen. Figure represents an example of WGM resonance wavelength shifts ($\Delta\lambda$), that denotes the binding event of 3PGK to the sensor. Alongside the changes of $\Delta\lambda$, the changes of the full width at half maximum, $\Delta\kappa$, of the WGM were registered. At relatively low intensities of WGM, binding events show red-shifts of λ, and do not show the changes of $\Delta\kappa$. Increasing WGM intensities led to observations of single-molecule detection as close to zero change of λ accompanied by changes of $\Delta\kappa = 3$ fm on average. We consider that this mechanism is related to the strong mutual influence of WGM

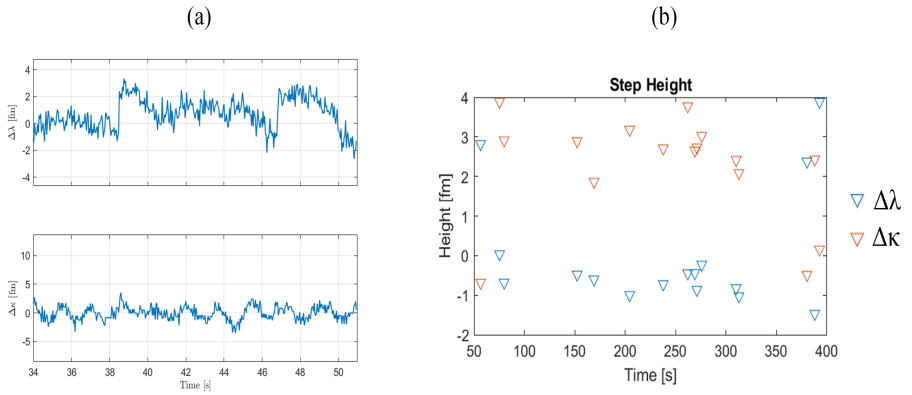

Fig. 1. (a) Traces of WGM resonances: Δλ (top) and Δκ (bottom) vs time – low WGM intensity. (b) Values of Δλ and Δκ – high WGM intensity.

(a) Traces of WGM resonances: $\Delta\lambda$(top) and $\Delta\kappa$(bottom) vs time – low WGM intensity. (b) Values of $\Delta\lambda$ and $\Delta\lambda$ – high WGM intensity

modes and plasmon nanorods, where nanorods form hot spots due to plasmon-enhanced fields. At the same time, they generate increased temperature due to Ohmic losses originating from plasmonic oscillations. Binding events of 3PGK molecules to the nanorod, usually causing red-shifts, are compensated due to partial absorption of optical energy by different parts of the molecule and partially due to the absorption of the excessive heat energy. In turn, the absorbed energy and temperature make the refractive index smaller in the hot spots, resulting in a compensating resonance wavelength shift. Thus, we demonstrated a new sensing mechanism, which is caused by thermal changes in optoplasmonic sensors.

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Fiber Fabry-Pérot micro-resonators for molecular oxygen A-band determination using modulation spectroscopy

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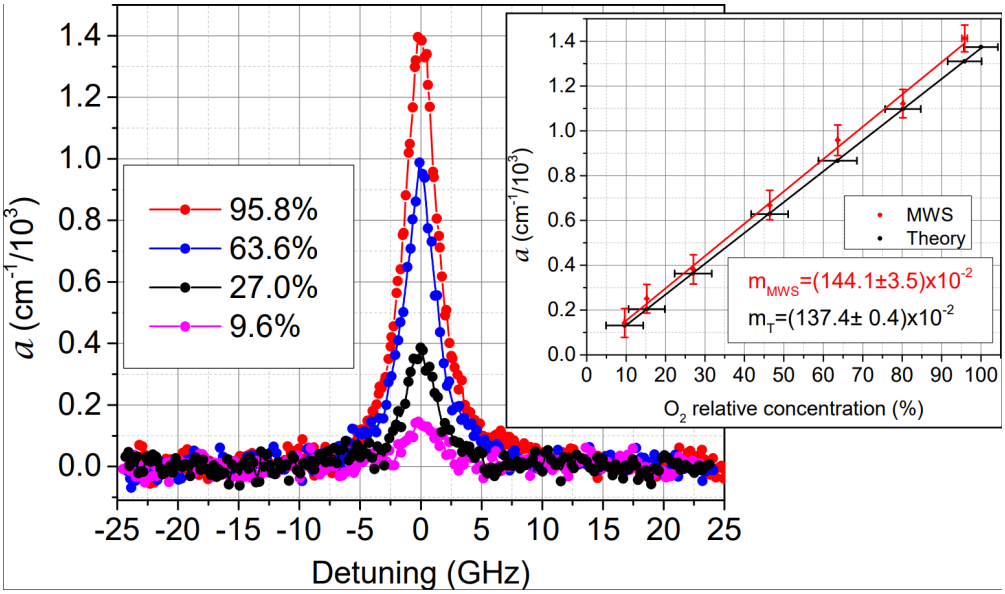
Fabry-Pérot cavities has been used extensively for the study of different atmospheric gas species as they can provide fast and precise observations of their spectrum [1, 2]. Miniaturized detection systems as the monolithic fiber Fabry-Pérot micro-cavities [3] can offer a compact, lightweight and robust alternative for onsite experiments realization using standard techniques such as cavity mode-width spectroscopy, demonstrated here. Furthermore, fast acquisition techniques using cavity modulation spectroscopy can exploit the noise profile of the cavity system to resolve the oxygen A-band, with higher resolution.



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A silica Rib microdisk for visible comb spectroscopy

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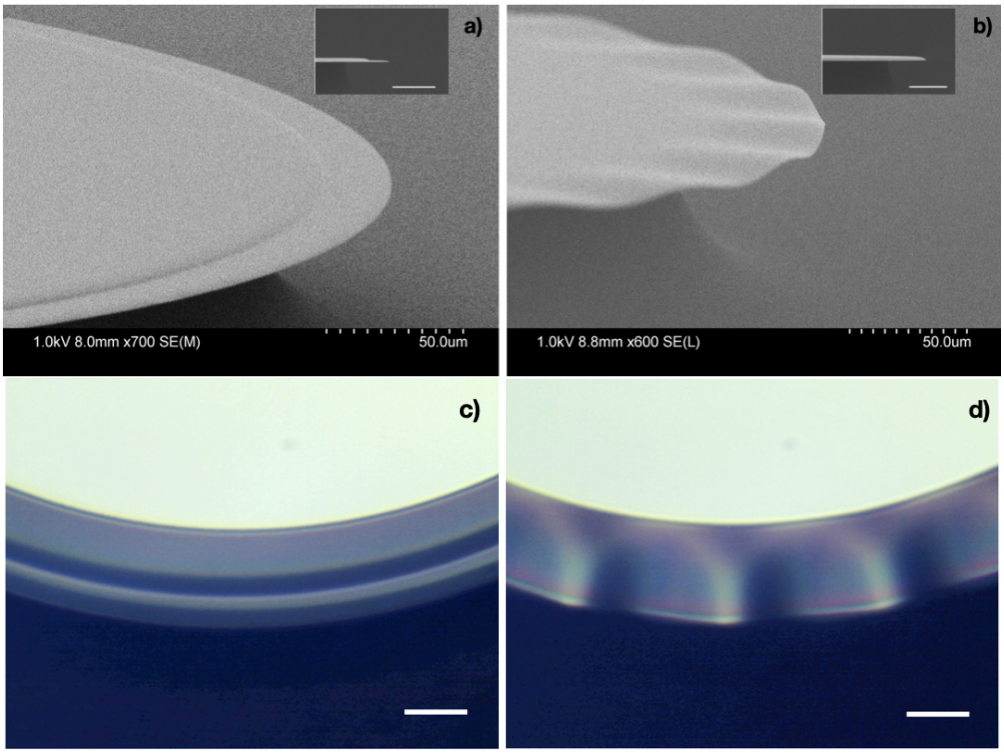
Thin silica microdisks with sub-micron thickness are essential for soliton visible comb generation, due to the need for dispersion engineering at such wavelengths [1]. Furthermore, large disk size is desirable to explore the complex spectral structure of bio-molecules since the density of the comb would be higher in disks. Hence, the need for a fabrication process to make large thin microdisk is of great importance for ongoing research toward soliton visible comb generation for precision in-vitro spectroscopy of single bio-molecules. Thermal stress induced buckling has been one of the limiting



factors on silica microdisk fabrication [2]. This residual stress arises from the difference in thermal expansion coefficient of thermally grown silica and the silicon substrate, when cooling down from growth temperature (exceeding 1000 °C). This stress at the interface, will then release upon the disk fabrication, and causes the edges of the microdisk to buckle. The buckle effect significantly reduces the optical quality factor the disk, making the comb generation infeasible. This mechanical instability is more severe in thinner and larger disks, which are necessary for dense visible comb generation. Here, we report a new silica microdisk geometry to overcome the buckling challenge in such disks. The thick top part of the disk as shown in Ffigure a) and figure c) provides the mechanic strength to prevent the disk from buckling while the submicron thin rib at the edge of the disk can provide proper dispersion for soliton generation at visible wavelengths. The rib-shaped geometry effectively solved the buckling problem in 1 mm disks with sub-micron thickness as evident by comparing to a conventional disk of the similar size, thickness and undercut shown in the subplots of figure b) and figure d). Furthermore, optical quality factors as high as 12 million was achieved at 970 nm wavelength which will be sufficient for comb generation.

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Adding Quantum Confinement to Cavity Molecular Polaritons

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By applying lateral confinement in an optical cavity, we extended the "particle-in-a-box" idea, which was broadly developed in semiconductor quantum dot research, into mid-Infrared (IR) Fabry-Perot cavity modes. The quantized cavity modes hybridize with molecular vibrational modes, resulting polariton states that can support multiple coherence states in the IR regime. We applied tailored pump pulse sequences to selectively prepare these coherences and collected 2D IR spectra to verify their existence. We found that because the polariton quartet reside in the same cavity, they were specifically robust towards phase fluctuations in space. The multiple robust coherences of the confined cavity polaritons pave the road for entangled states and manipulation of interactions between states in different cavities for future quantum simulation using vibrational polaritons.

Fiber optic strain sensing with π-shifted Fiber Bragg Gratings

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CT

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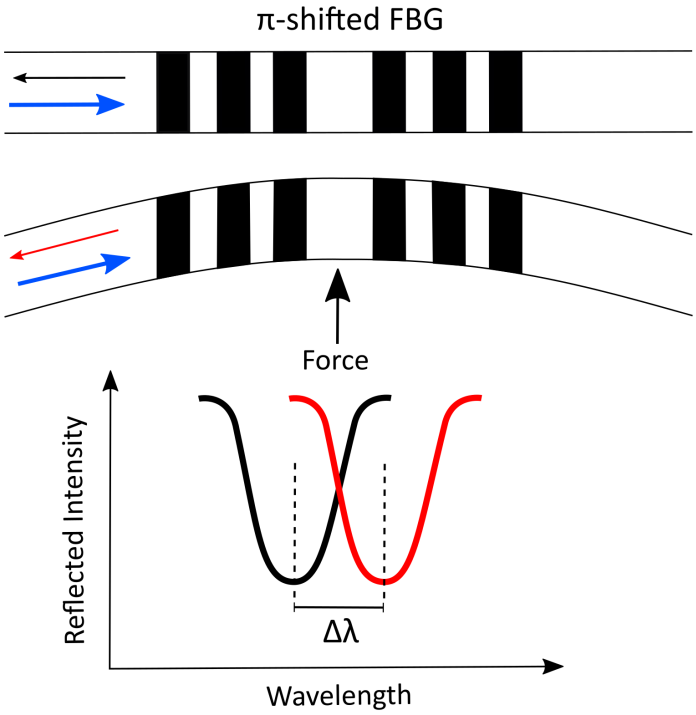
Optical strain sensing is an emerging field of research in recent years due to its vast applications in geotechnical monitoring and seismic sensing. Fiber-optic strain gauges, unlike conventional electrical strain gauges, are passive sensors which are immune to electromagnetic interference (EMI). This permits the sensor to be used in harsher environments, such as the ocean floor, but often requires the sensor to work over long fiber tether lengths. Previously this has been achieved using distributed sensing measurements which rely, typically on Brillouin scattering to detect and localize strain [1]. Highly localized or point sensing is more appropriate for acoustic (hydrophone)



measurements and submarine equipment monitoring and can be achieved using a compact transducer, typical a Fiber Bragg Grating (FBG) or equivalent, which exhibit a resonance peak/dip in the reflection spectrum when interrogated by a laser. This peak shifts linearly when strain is applied to the grating. Our group had previously demonstrated strain measurements up to 10 km with a fiber Fabry-Perot interferometer (a pair of identical FBGs) but were limited by noise and lock instability due to Stimulated Brillouin Scattering (SBS) and polarization-dependent peak splitting [2].
Here we present a strain sensing system capable of measuring strain over 50 km with the use of a π-shifted FBG and an EDFA. The broader-linewidth peak of π-shifted FBG combats the peak splitting and the use of an EDFA to amplify the reflected beam greater improves the signal to noise ratio over large tethers. The strain is measured using the Pound-Drever-Hall method to lock the laser to the center of the reflected resonance and observe the feedback signal required to maintain the lock. The system was locked through 50 km of fiber and strain was measured at 10 Hz with a

sensitivity of 10.65 mV/$\mu\varepsilon$. This system has promising applications for monitoring seismic activity by using the sensor head in a hydrophone in place of the conventional piezoelectric strain gauge.

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List of Posters

Tuesday Session

Fabrication of tunable fiber Fabry-Pérot microcavities

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We report on our recent development of Fiber Fabry-Pérot cavities (FFPCs). FFPCs offer higher finesse, reduced mode waist, lower mode volume, and better mode matching with respect to other macroscopic cavities. These advantages increase the potency of light-matter interactions within these microcavities. One can harness the exquisitely sensitive open-access geometry of the FFPC to eliminate many experimental compromises and implement new architectures for applications such as relaying quantum information, spectroscopically sensing single molecules, or influencing reaction chemistry. We present results on the formation of the concave fiber mirrors that form these cavities. Treating the tip of an optical fiber with a high-energy CO_2 laser pulse forms an ultra-smooth concave mirror surface. By carefully manipulating the spatial profile, pulse duration, and power density of the laser beam, we can modify the shape of the ablation.

Single-adenylate kinaseturnover events studied via WGM microcavities

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[2]University of Bath, Bath, UK

Whispering-Gallery Mode (WGM) biosensors are in development for the detection of single small-molecules, biophotons and enzyme dynamics. Understanding protein mechanisms, including their dynamics, is of huge importance for biotechnology and treating disease. An area of current interest regards conformational change and how these promote catalysis and communicate allostery. However, investigations into these subjects are complex, often limited due to short molecular dynamics simulation time periods, addition of molecular fluorescent tags, or time-consuming and expensive isotope labelling. The coupling of WGM microcavities to plasmonic nanoparticles, with its greater sensitivity, will allow detection of minute changes of enzyme polarizability in the enhanced near-field of a nanorod, of which is dependent on its conformation and orientation on the sensor. This allows label/tag-free single-molecule investigations of native conformational changes during reaction cycles and interactions with allosteric regulators.This investigation initially focussed on observing the reversible turnover of adenylate kinase (Adk). Adk was immobilised on a gold nanorod-glass sphere microresonator via a C-terminal cysteine. The microcavity construct was coupled to the evanescent field of a 780 nm beam in a prism and immersed in buffer containing substrates of the forward or reverse reaction. 3-phosphoglycerate kinase (3PGK) was used in previous investigations for a similar goal, producing double-peaked signals at intervals similar to bulk-measured kcat. Due to PGK-Adk overall structural similarity, we predicted a similar signal architecture. Results confirmed a resemblance in signals was observed between Adk and 3PGK, both in the forward and reverse reactions.To understand how this signal architecture can change, a common Adk inhibitor, Ap5A, was added to the system. Previous NMR experiments show Ap5A binding is slow [1], and therefore we predicted that the signal architecture would be elongated over a greater time period. Longer signals periods were indeed observed, possibly showing the formation of the enzyme-inhibitor complex, and therefore the “closed” conformation. These results demonstrate proof of concept: WGM biosensors can detect conformational changes at nm level, opening the door to a plethora of investigations into enzyme functionality.

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Broadband Mid-infrared Spectroscopy for plasma analysis

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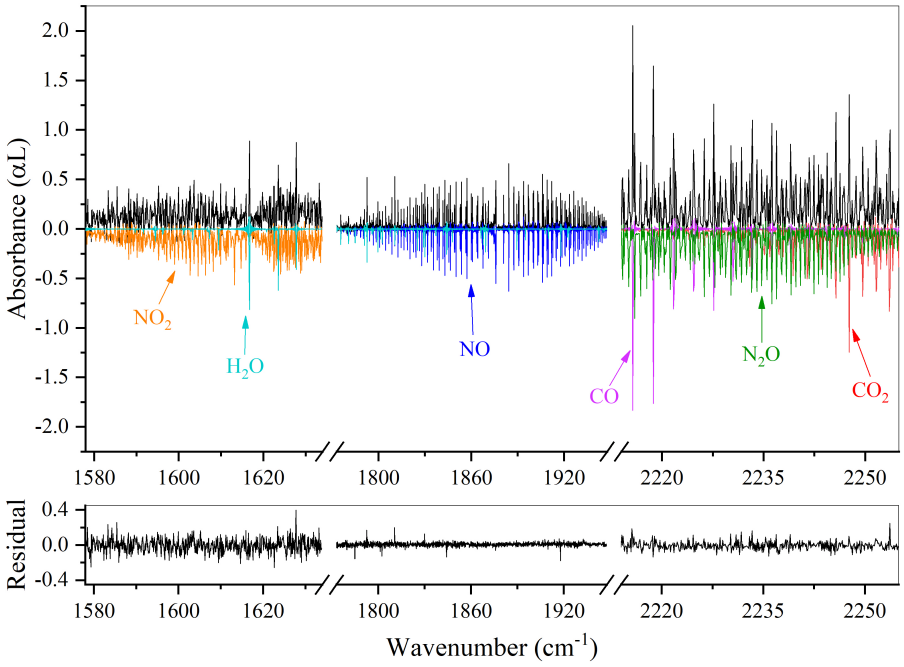
[2]Technical University of Denmark, Lyngby, Denmark
[3]Maastricht University, Maastricht,The Netherlands

Recycling the greenhouse gases by an electrified chemical process using green energy is of particular interest in recent years. For instance, plasma-based gas conversion, is able to transform efficiently carbon dioxide (CO_2) and methane (CH_4) into carbon monoxide (CO) and hydrogen (H_2), also known as syngas [1]. However, discharge plasmas consist of numerous (reactive) species, and it is challenging to control the reaction specificity. Therefore, it is of great importance to develop a multispecies detection system capable of analyzing the reactants/products of the plasma for different conditions. Ultra-broadband spectroscopy in the mid-infrared (MIR) wavelength range, where most of the molecular species have their distinct and strong absorption features, has a great potential to be used for this purpose. We have recently demonstrated the potential of the a



home-built Fourier Transform Spectrometer based on a novel MIR supercontinuum (SC) source [2] and a multipass absorption cell for multispecies trace gas detection [3]. Here, we use this system to monitor the reactants/products of CO_2/CH_4/N_2 discharge plasmas. For instance, the figure below shows the measured absorbance spectrum of the product gasses (in black) in the outflow of a discharge plasma of 50% CO_2 in N_2, alongside the corresponding fitted simulated spectra (inverted, in color) of NO_2 (~ 154 ppm), NO ($\sim 0.33\%$), N_2O (~ 570 ppm), CO ($\sim 14.6\%$), and CO_2 ($\sim 13.0\%$). These measurements demonstrate the ability of the system to detect numerous molecular species created in the plasma, even for the species with overlapping spectral features. By changing the parameters of the plasma, such as specific energy input and reactant mixing ratio, one can find the best combination of the parameters for maximizing the concentration of a specific produced species.

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Additional Links

[1]R. Krebbers, N. Liu, K. E. Jahromi, M. Nematollahi, O. Bang, G. Woyessa, C. R. Petersen, G. van Rooij, F. J. M. Harren, A. Khodabakhsh, and S. M. Cristescu, "Mid-infrared supercontinuum-based fourier transform spectroscopy for plasma analysis", Scientific Reports **12**, 9642 (2022).

Mid-Infrared frequency comb spectroscopy for breath analysis

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Mid-infrared (MIR) comb spectroscopy in the chemical and biological fingerprinting region ($2.5 - 25\ \mu$m) [1, 2] can enable a new regime of health diagnostics [3]. Specifically, cavity-enhanced frequency comb spectroscopy covering $\sim 1.5 - 1.7\ \mu$m [4] and $\sim 3 - 4\ \mu$m [5, 6] has been used to measure chemicals in human breath at parts-per-trillion and parts-per-billion (ppb) concentrations. However, a frequency comb spectrometer with full coverage from $3 - 12\ \mu$m is desirable to measure a range of biomarkers linked with multiple health conditions [3], while preserving high-resolution and ppb sensitivity. MIR frequency combs produced from intrapulse difference frequency generation (IP-DFG) [1, 2] can provide improved bandwidth and resolution, while pathlength enhancement and coherent averaging from dual-comb spectroscopy can increase detection sensitivity.

We investigate pathlength-enhanced MIR dual-comb spectroscopy in the fingerprinting regime. We generate MIR frequency combs via IP-DFG by focusing a near-infrared comb into a nonlinear crystal. As volatile organic compounds at $\sim 10 - 250$ ppb concentration in human breath have been linked with early active viral infections [7], we optimize the MIR comb spectrum to measure their distinguishing spectral features simultaneously from $3 - 10\ \mu$m. We employ an astigmatic Herriot cell aligned for an ~ 85 m path enhancement to improve sensitivity to ppb gas concentrations.

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Cavity ring-down spectroscopy of small hydrocarbons in hypersonic Laval expansions

Eszter Dudas[1]**,** ***Lucile Rutkowski***[1]**,*****Samir Kassi***[2]**,** ***Christine Charles***[3]**,** ***Robert Georges***[1]

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[2]University of Grenoble Alpes, CNRS, LIPhy, Grenoble, France
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Inversion of space telescope observation requires reference laboratory spectra at temperatures relevant to astrophysical environments. In particular, line by line assignment of the hot band transitions of hydrocarbons are of importance for the inversion of exoplanets observations. We will introduce the SMAUG apparatus (Spectroscopy of Molecules Accelerated in Uniform Gas flows, [1]) developed to study rotationally-cold hot-bands. It relies on cavity-ring down spectroscopy (CRDS) in a hypersonic jet environment. We will show its application to the highly excited states of methane and the beginning of the analysis of the cold bands of ethylene. We detect many rotationally resolved transitions and use them to improve the TheoReTS spectroscopic database [2]. This is expected to bring numerous insights in the applications looking for a quantitative, multispecies and sensitive instrument, such as high temperature spectroscopy for line assignments.

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Ring Cavity OF-CRDS spectrometer based on SG-DBR lasers

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A spectrometer based on optical-feedback cavity ring-down spectroscopy (OF-CRDS) has been developed with a tunable sampled grating distributed Bragg reflector (SG-DBR) laser and a ring resonant optical cavity. Shot-to-shot variations in the ring-down decay constant were as low as 15 ppm.

Cavity ring down supersonic jet spectrometer based on a MIR cw-OPO Laser

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There is a surprising chemical diversity of molecules seen in space [1]. In the vicinity of old dying stars, atoms combine to form molecules and serve as seeds for dust formation. In many cases the cirumstellar molecules are, unlike here on earth, unsaturated compounds, i.e. radicals, or of ionic nature. These species, which are difficult to produce on earth, are valuable probes to determine the physical conditions in stellar envelopes [2].
Our aim is to perform spectrally resolved and extremely sensitive measurements on astrophysically relevant molecules in the mid-infrared region utilizing the cavity ringdown (CRD) method. Our new experimental setup is a combination of an AOM switched „Scanned Cavity" MIR CRD spectrometer with a supersonic jet molecule source. The CRD mirrors have a $R > 99.99\%$ between 3.0 and $3.4\,\mu$m. In this region, O−H, N−H and C−H stretching vibrations can be probed. First calibration measurements on weak bands of N_2O will be presented. Our setup can be combined with an electrical discharge source and a heating source (e.g. for hydrogen peroxide), to investigate spectra of yet unknown transient species.

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Fourier transform and grating-based spectroscopy for multi-species trace gas detection utilizing mid-infrared supercontinuum source

Khalil Eslami Jahromi, Mohammadreza Nematollahi, Roderik Krebbers, Amir Khodabakhsh, Simona M. Cristescu, Frans J. M. Harren

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We present a multi-species trace gas sensor based on a fast and compact home-built Fourier transform spectrometer (FTS) and a scanning grating-based spectrometer (GS), both combined with a multi-pass cell and a broadband mid-infrared supercontinuum (SC) source.
Fourier transform infrared (FTIR) spectroscopy is a well-established scientific method widely used for broadband molecular spectroscopy. The FTIR spectrometer can support both broad spectral coverage (required for multi-species gas detection) and high spectral resolution with a calibrated frequency scale. Traditionally, FTS employs an incoherent and omnidirectional thermal source with low brightness output. This restricts exploiting an optical cavity or a multi-pass cell to achieve a long absorption path length for gas-phase spectroscopy. Combining advanced reliable MIR SCs with FTS has circumvented these challenges. They offer high spatial coherence and a very high spectral brightness, much higher than thermal sources.
Conventionally, GSs are also very popular due to the low price and ease of use. They can generally provide enough spectral resolution to be captured by a fixed linear camera or alternatively a single-point detector while scanning the grating.
The FTS covers the spectral bandwidth of the SC source ($2-4\,\mu$m) and provides a best spectral resolution of 1 GHz in 6 seconds. The GS support a spectral range of $3.1-3.7\,\mu$m with 75 GHz resolution in 100 ms. Both spectrometers have a detection sensitivity of a few hundred of ppbv/$\sqrt{Hz}$ for different gas species. We evaluate the performance of the GS and FTS-based sensors for quantifying gas concentrations in a complex mixture of various gases. In addition, we measure fruit-produced volatiles which are gas biomarkers for fermentation (ethanol, acetaldehyde, ethyl acetate), ripening (ethylene), and rotting (acetone, methanol). Both sensors are suitable for various gas sensing applications, and their selection depends on the requirements of a specific application.

Towards whispering-gallery mode Laser realisation with fluorophores for biosensing applications

Samir Vartabi Kashanian, Frank Vollmer, Deshui Yu

University of Exeter, Exeter, UK

We present a multi-species trace gas sensor based on a fast and compact home-built Fourier transform spectrometer (FTS) and a scanning grating-based spectrometer (GS), both combined with a multi-pass cell and a broadband mid-infrared supercontinuum (SC) source.
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Cavity enhanced chiral optical rotation

Michail Xygkis[1,2]**,** ***Michail Koutrakis***[1,2]**,** ***Kostas Tazes***[1,2]**,** ***Georgios E. Katsoprinakis***[1]**,** ***T. Peter Rakitzis***[1,2]

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Chiral sensing plays a crucial role in numerous fields of science and industry. The measurement of weak chiral signals is limited by spurious birefringence, as well as slow and imperfect background subtraction procedures. We present a novel chiral cavity-enhanced polarimeter (CCP), an optical instrument in which the sensing light traverses the chiral sample multiple times, thus amplifying the weak chiral optical rotation signal by the number of cavity round-trips (typically > 100). The hallmark of this technique is the implementation of rapid signal reversals, which eliminate the need for background subtractions, and enable absolute chiral measurements even in noisy environments, resulting in greatly enhanced sensitivities (~ 0.02 mdeg·Hz$^{-1/2}$), relative to commercial instruments (~ 5 mdeg·Hz$^{-1/2}$).

High-accuracy measurement of Mid-IR refractive indices of GaAs/AlGaAs in thin-film multilayers

***Lukas W. Perner*[1], *Georg Winkler*[1], *Gar-Wing Truong*[2] *David Follman*[2], *Jakob Fellinger*[1], *Maximilian Prinz*[1], *Stephan Puchegger*[1], *Garrett D. Cole*[2], *Oliver H. Heckl*[1]**

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Thin film optical coatings are essential for a wide range of photonic applications. However, meeting design targets without resource-intensive iterations in fabrication relies on precise knowledge of the optical properties of the deposited thin films, especially their refractive index. Accurate knowledge of the refractive index for GaAs-based materials is required for the design of a plethora of optical devices in the near- (NIR) and mid-infrared (MIR) spectral region, among them vertical-cavity surface-emitting lasers [1] and high-performance substrate-transferred optical interference coatings [2].
Typically, the refractive index shows variability depending on deposition conditions, and are therefore often insufficiently accurate [3]. This motivates the need for routine characterization of as-produced thin film structures. While several measurement methods for refractive indices exist – among them (spectroscopic) ellipsometry, the prism method, and Fourier-transform Infrared (FTIR) refractometry [3] – they all rely on specialized samples and measurement equipment.
We present a novel, robust method to measure the refractive indices of transparent materials over the NIR and MIR spectral regions, involving the following steps: Obtaining a photometrically-accurate low-resolution transmittance spectrum of a quarter-wave-type GaAs/AlGaAs structure via FTIR; extraction of individual layer thicknesses from cross-sectional imaging of the structure via calibrated scanning electron microscopy; using these thickness values, together with a single effective oscillator model [4] for the refractive indices, to fit a transfer-matrix-method model to the FTIR spectrum. Uncertainty propagation is done via a Monte-Carlo-type propagation method, including uncertainties in both, layer thicknesses and FTIR, measurements.
This method allows us to simultaneously extract the refractive indices of both materials in the as-deposited thin film structure with per-mille-level relative uncertainty over the $2-7\ \mu$m range.

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