

Users Meeting of Cavity Spectroscopy, 18 December 2025, RIKEN, Japan

Area	US	US	US	Euro	China	Japan							
Time zone	PST	MST	EST	CET	CST	JST							
Time difference from UTC	-8	-7	-5	1	8	9							
day	h	h	h	h	h	h	m	Session	No.	Ivent	Type	Duration	Chair
2025/12/16													
2025/12/17	17	18	20	2	9	10	0			Zoom Test (1 hour)			
2025/12/18	16	17	19	1	8	9	30			Site opening Zoom Test (30 min) PowerPoint file copy for in-person			
2025/12/18	17	18	20	2	9	10	0	Session 1	1-1z	Opening remarks			Araki
							5	(English)		Qizhong Liang (Invited)	Zoom	25 minutes	Iwakuni
							10						
							15						
							20						
							25						
							30	1-2z		Qi Huang		15 minutes	
							35						
							40						
							45	1-3z		Jinke Li		15 minutes	
							50						
							55						
	18	19	21	3	10	11	0	1-4		Mitsunori Araki	in person	25 minutes	
							5						
							10						
							15						
							20						
							25	1-5		Garrek Stemo		15 minutes	
							30						
							35						
							40			PowerPoint file copy for in-person			
							50			Lunch (1 hour)			
	19	20	22	4	11	12	0			↓			
							50			RIKEN sightseeing			
	20	21	23	5	12	13	0			↓			
							20			Zoom Test (30 min)			
							25			PowerPoint file copy for in-person			
							30			Short anounce			
							35	Session 2	2-1	杉本良介 (Invited)	in person	25 minutes	Oyama
							40	(Japanese)		Tryosuke Sugimono			
							45						
							50						
							55						
	21	22	0	6	13	14	0	2-2		星野翔麻		25 minutes	
							5			Shoma Hoshino			
							10						
							15						
							20						
							25	2-3		岩國加奈		25 minutes	
							30			Kana Iwakuni			
							35						
							40						
							45						
							50	2-4		小谷隆行		15 minutes	
							55			Takayuki Kotani			
	22	23	1	7	14	15	0						
							5	2-5z		阿部恒	Zoom	15 minutes	
							10			Hisashi Abe			
							15						
							20			Cofee Break			
							30			Zoom test from Europe			
							55			Short anounce			
	23	0	2	8	15	16	0	Session 3	3-1z	Vittorio D'Agostino	Zoom	25 minutes	Araki
							5	(English)					
							10	from					
							15	Europe					
							20						
							25	3-2z		Lucile Rutkowski		25 minutes	
							30						
							35						
							40						
							45						
							50	3-3z		Anthony Roucou		25 minutes	
							55						
	0	1	3	9	16	17	0						
							5						
							10						
							15			Closing remarks			Araki
							20			Photo			
							0			Moving to the reception venue			
	1	2	4	10	17	18	30			Reception			
	3	4	6	12	19	20	30			Reception closing			

1-1z.

Breath Analysis by Modulated Ringdown Comb Interferometry

Qizhong Liang

University of Colorado Boulder

Breath analysis examines the gas molecules exhaled from breath and associates their change in concentration with medical conditions in a non-invasive manner. Using cavity-enhanced frequency comb spectroscopy—a broadband laser absorption spectroscopy technique—we demonstrated COVID-19 breath detection with 85 % accuracy in a 170-subject cohort. An even better diagnostic performance would require us to detect even more breath molecules. However, the extension of spectral coverage and cavity finesse to detect molecules of different chemical bonds and of lower concentrations has been a long-standing challenge, bottlenecked by the cavity-comb frequency mismatch introduced by the intracavity dispersion. Here we resolve this challenge with Modulated Ringdown Comb Interferometry, a dispersion-immune technique enabling high-throughput and robust measurement of photon decay rates at massively parallel discrete optical frequencies. This technique has allowed us to establish a new gas sensing record with a 5 km absorption path length and 1,010 cm⁻¹ mid-infrared coverage, with which we demonstrated simultaneous measurement of 20 molecular species at above 1-part-per-trillion sensitivity varying in concentrations by seven orders of magnitude.

1-2z.

Cavity-Enhanced Doppler-Broadening Thermometry via All-Frequency Metrology

Qi Huang, Jin Wang, Rui-Heng Yin, Yan Tan, Cun-Feng Cheng, Yu R. Sun, An-Wen Liu, and Shui-Ming Hu

University of Science and Technology of China

We demonstrate Doppler broadening thermometry (DBT) with all-frequency-domain measurements. Using the R(10) transition of CO at 1567 nm in a high-finesse optical cavity (mode width 0.6 kHz), we resolve Doppler profiles with high signal-to-noise ratios across 2 - 17 Pa pressures. A global Voigt-profile analysis yields temperatures deviating by only -2.0 ± 3.6 mK from calibrated thermometers, with systematic errors suppressed below 9 ppm. The results show negligible dependence on lineshape models when accounting for pressure effects, resolving a long-standing challenge in DBT. This approach establishes a new paradigm for quantum-based thermometry and provides a precision platform for testing molecular collision physics.

1-3z.

Precision Spectroscopy and Line-Intensity Ratio Thermometry: Breaking Barriers in Metrology

Jin-Ke Li, Jin Wang, Rui-Heng Yin, Qi Huang, Yan Tan, Chang-Le Hu, Yu R. Sun, Oleg L. Polyansky, Nikolai F. Zobov, Evgenii I. Lebedev, Rainer Stosch, Jonathan Tennyson, Gang Li, and Shui-Ming Hu

University of Science and Technology of China

Accurate determination of molecular transition intensities is vital to quantum chemistry and metrology, yet even simple diatomic molecules have historically been limited to 0.1% accuracy. Here, we show that frequency-domain measurements of relative intensity ratios outperform absolute methods, achieving 0.003% accuracy using dual-wavelength cavity mode dispersion spectroscopy. Enabled by high-precision frequency metrology, this approach reveals systematic discrepancies with state-of-the-art ab initio calculations, exposing subtle electron correlation effects in the dipole moment curve. Applied to line-intensity ratio thermometry (LRT), our technique determines gas temperatures with 0.5 millikelvin statistical uncertainty, exceeding previous LRT precision by two orders of magnitude. These results redefine the limits of optical gas metrology and enable International System of Units traceable measurements for applications from combustion diagnostics to isotopic analysis. Discrepancies of up to 0.02% in transition probability ratios challenge theorists to refine models, establishing intensity ratios as a paradigm in precision molecular physics.

1-3.

Polarization Dependence of Super Mirror for Cavity Ringdown Spectroscopy: Results Report

Mitsunori Araki and Kohsuke Suma

Tokyo University of Science, Kagoshima University

A long optical path length is the most critical factor in achieving high sensitivity in spectroscopy. In the case of cavity ringdown spectroscopy, a cavity consisting of two supermirrors provides a long path length, with the high reflectance of the mirrors arising from their low excess loss. For mirrors coated with crystal, the excess loss has been reported to depend on polarization. However, an amorphous coating supermirror was expected to show a negligible polarization dependence. In this work, we measured the excess loss as a function of mirror rotation around its optical axis in the optical region at 681.2 nm for the three amorphous coating supermirrors produced simultaneously by vapor deposition in the same furnace. The backside mirror of the cavity was rotated, and the ringdown time as a function of rotational angle was measured at 10-degree intervals. We observed sinusoidal variations in excess loss by rotation. The most considerable difference in excess loss during the rotation of the back mirror reached a maximum of 5.8 ± 1.2 ppm. This difference demonstrates the importance of optimizing the rotational angle alignment of mirrors to achieve high sensitivity via a long path length.

1-4.

Exploring the Rabi splitting parameter space in vibrational polaritons

Garrek Stemo, Avram Gutierrez, Joel Nishiuchi, Masaki Kiyokawa, and Hiroyuki Katsuki

Nara Institute of Science and Technology, Division of Materials Science

Investigations into molecular polaritons, formed via strong interaction between a cavity optical mode and material transition, continue to deliver tantalizing suggestions of their potential to systematically manipulate chemical and quantum properties of molecular ensembles. Experiments have demonstrated modified chemical reactivity, material properties, and energy transfer. Mechanisms remain elusive, but ultrafast pump-probe spectroscopy is the best tool to study the microscopic processes that lead to the observed macroscopic molecular modifications.

Very recently, several reports on polariton dynamics have shown that nearly all late-time behavior can be explained by dark state molecular population dynamics (after polariton dephasing). This idea has only been experimentally investigated in two standard samples with relatively small Rabi splitting values, a measure of the light-matter coupling strength, and primarily at resonance conditions. We systematically explore this parameter space using a sample capable of values three times larger than reported so far. By sweeping both magnitude (via molecular concentration) and energetic detuning between the cavity mode and molecular vibrational energy, we analyze subtle changes in nonlinear spectral features and time-resolved behavior. Our analysis reveals the extent to which polaritons can act as reporters of underlying dark state behavior and the interplay between molecular and polaritonic behavior.

2-1.

光共振器を用いた宇宙重力波検出器に向けたバックリンク干渉計の実証

Demonstration of a Back-Link Interferometer for Space-Based Gravitational-Wave Detectors with Optical Cavities

杉本良介／東京大学

時空の歪みの波動現象である重力波の観測は様々な知見をもたらす。特に現在稼働している地上検出器では観測が困難な 10 Hz 程度よりも低い周波数帯は究極的には初期宇宙からの原始重力波など様々な成果が期待される。その観測に向けた有望な手段の一つとして各宇宙機間で光共振器を構築する光共振器型宇宙検出器がある。中でも光共振器に対して周波数をロックした 2 つのレーザー間の差周波から重力波信号を抽出するバックリンク干渉計と呼ばれる方式は各宇宙機間の相対距離の精密制御が不要という利点がある。一方、課題としてレーザー周波数雑音による感度の悪化があり、この解決のため取得した信号から事後的な周波数雑音の“引き算”が理論提案されていた。本講演では高感度化の達成に必須となる周波数雑音の引き算に関する初の実証結果について PRD.109.022003 で出版した内容を基に紹介する。

2-2.

時間分解キャビティリングダウン分光によるイソプレン由来クリーギー中間体とカルボン酸の反応速度の決定

Determination of Reaction Rate between Isoprene Derived Criegee Intermediate and Carboxylic Acids by Time-Resolved Cavity Ringdown Spectroscopy

星野翔麻（北里大学理学部化学科）、秦寛夫、吉越裕介、

戸野倉賢一、松木亮、石川春樹、築山光一

Criegee 中間体 (CI) は大気中の揮発性有機酸化物 (VOC) のオゾン分解反応の過程で生成するカルボニル酸化物 (R_1R_2COO) であり、分子科学および大気化学的な注目を集めている反応活性種である。近年では、大気中の CI の主要な失活経路として、蟻酸などの有機酸との反応の重要性が注目されている。植物の代謝過程で大気中に放出されるイソプレンは、大気中の VOC の 60 ~ 90 % を占めると考えられており、したがってイソプレン由来の CI の反応性を明らかにすることは極めて重要である。本研究では、時間分解型キャビティリングダウン分光法 (CRDS) と量子化学計算/遷移状態理論を用いて、イソプレン由来の CI であるメチルビニルケトンオキシド (MVKO: $CH_3C(C_2H_3)OO$) とカルボン酸との反応による不揮発性有機化合物生成の反応速度および反応機構を解明した。さらに得られた速度定数を全球化学輸送モデルに組み込み、MVKO とカルボン酸の反応による大気中のカルボン酸の消失割合を試算した。

2-3.

共振器分光ことはじめ

An Introduction to Cavity Spectroscopy

岩國加奈／電気通信大学

光共振器を分光セルとして用いると高感度あるいは高分解能な測定が可能となり、この特性はこれまでのキャビティ分光ユーザーズミーティングで紹介されたように様々な研究に活用されている。しかしながら、共振器の設計や構築にはノウハウが必要であるため初めて取り組むにはハードルが高く、それに関わらず学術論文には詳細の記述は省かれる。本講演ではこれから共振器分光の導入を検討する方に向けて、事前に考慮すべき点や実験の最初の一步目から二歩目あたりで必要となる事項について紹介する。

2-4.

太陽系外惑星大気の詳細観測に向けたキャビティ分光による分子データベースの構築

Building a Molecular Database for Detailed Observations of Exoplanet Atmospheres Using Cavity Spectroscopy

小谷隆行（アストロバイオロジーセンター）、岩國加奈、河原創、細川晃

太陽系外惑星は 1995 年に初めて発見され、現在では 6000 個以上が確認されている。近年、分光観測技術の進展により、系外惑星大気中に多様な分子が検出されてきた。しかし、高分散スペクトルと既存の分子データベースの整合性に問題があり、メタンなど重要分子の解析が困難になっている。この不整合は、今後発見される低温～温暖な木星型惑星、いわゆる木星の姉妹惑星の反射光スペクトル解析において深刻な障害となる。我々はこの課題を解決するため、惑星と同条件の温度・圧力下で模擬大気を用い、キャビティ分光法によってメタン吸収線を測定する計画を進めている。得られたデータを基に分子ライナリストを構築・校正し、宇宙望遠鏡や地上大型望遠鏡による低温・温暖木星型惑星の正確な物理量推定に貢献することを目指す。

2-5z.

レーザー波長同調型キャビティリングダウン分光法の開発

Development of laser wavelength-tuned cavity ring-down spectroscopy

阿部恒／産総研

キャビティリングダウン分光法(CRDS)は、レーザー吸収分光法の一つであり、微量ガスの計測、特に微量水分の測定に威力を発揮する計測法である。高フィネス光共振器（キャビティ）を用いることで、数十 km に及ぶ有効光路長を確保し、高感度化を実現する。さらに、吸収係数の絶対値が得られるため、標準ガスを使用せずに定量測定が可能となる。CRDS では、プローブレーザーの周波数をキャビティの共振周波数に一致させる必要がある。本研究では波長計を用いてレーザー波長を目的の値に合わせた(同調した)状態で、キャビティの共振周波数を変化させてリングダウン時間(τ)の測定を行う「レーザー波長同調型 CRDS」を開発した。

3-1z.

Linear optical-feedback cavity ring-down spectroscopy at 3- μm

Vittorio D'Agostino, Emanuele Tofani, Stefania Gravina, Eugenio Fasci, Aniello Grado, Antonio Castrillo, and Livio Gianfrani

Dipartimento di Matematica e Fisica, Università degli Studi della Campania Luigi Vanvitelli, 81100, Caserta, Italy

We present a linear optical-feedback cavity ring-down spectrometer (LOF-CRDS) developed for high-sensitivity detection of light hydrocarbons under ultra-high vacuum (UHV) conditions. The system employs a 3 μm interband cascade laser (ICL), whose emission coincides with strong absorption features of the asymmetric C–H stretching vibrational mode. Passive optical feedback from the high-finesse linear cavity substantially narrows the ICL linewidth, ensuring efficient coupling into the resonator. An empty-cavity ring-down time of approximately 18 μs was measured, corresponding to a finesse of $\sim 37\,000$ and a cavity-mode width of ~ 9 kHz. The sensitivity of the spectrometer was evaluated through an Allan–Werle analysis at fixed laser frequency, from which we inferred a minimum detectable absorption coefficient of $\sim 2.5 \times 10^{-10} \text{ cm}^{-1}$ for a 100 s integration time. The spectrometer's performance was validated using the R(25) transition of the acetylene ν_3 band at $3352.2874 \text{ cm}^{-1}$, enabling a spectroscopic determination of the C_2H_2 partial pressure in the UHV chamber. For the selected line, the achieved sensitivity corresponds to a limit of detection of 2.1×10^{-8} mbar.

3-2z.

Direct frequency comb CRDS and CEAS in supersonic jets: pushing frequency comb-based cavity enhanced techniques toward cold reaction kinetics

Lucile Rutkowski, Romain Dubroeuq, Dominik Charczun, Piotr Maslowski, Robert Georges

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Kinetics of seemingly simple bi-molecular reactions remain challenging to study quantitatively at temperatures below 80 K, as it implies coupling advanced cooling techniques with time-resolved multi-species quantitative detection techniques. The interest toward direct frequency comb spectroscopy is rising steadily because of the technique selectivity, multispecies and quantitative abilities. However, sensitivity of such spectrometers remains limited, hence requiring enhancement using cavity techniques, for which frequency comb sources are well suited. I will present the current status of the experimental developments carried in our laboratory. In particular, we revisited the idea of using a time-resolved Fourier transform spectrometer to perform broadband cavity ring-down spectroscopy (CRDS, as introduced by Englert & Meijer in 1996) but implementing it with a fully stabilized near-infrared frequency comb (see Dubroeuq, and Rutkowski Opt. Exp 2022, and Dubroeuq et al. APL Phot. 2025). This opened up for precision spectroscopy of CO colliding with Ar, reaching sensitivity high enough to give evidence for speed-dependent effects in the absorption line profiles. Secondly, I will introduce our recent demonstration of near infrared comb cavity-enhanced absorption spectroscopy (CEAS) in a supersonic gas expansion (Dubroeuq et al. Molecules 2025), where the overtone absorption band of acetylene at 6K was recorded.

3-3z.

High-finesse cavity spectroscopy in the millimeter and submillimeter-wave range

Roucou, A., Simon, F., Elmaley, C., Pienkina, A., Chrayteh, M., Cuisset, A., Hindle, F., Mouret, G.
Université du Littoral Côte d'Opale, Laboratoire de Physico-Chimie de l'Atmosphère, France

The submillimeter-wave spectral range offers unique opportunities for molecular selectivity through rotational spectroscopy. However, the application of sensitive cavity-based techniques, well-established in the infrared, has historically been hindered by the lack of high-finesse resonators.

This work describes the operating principles and applications of high-finesse Fabry-Pérot cavities based on low-loss oversized corrugated waveguides and high-reflectivity photonic mirrors [1]. Achieving finesse values exceeding 3000, these setups provide effective interaction lengths of several kilometers in compact setups, enabling both Cavity-Enhanced Absorption Spectroscopy (CEAS) and Cavity Ring-Down Spectroscopy (CRDS). The instrument's capabilities are demonstrated through high-resolution sub-Doppler studies (Lamb-dip) and quantification of gases with limits of detection down to the parts-per-billion (ppb) level [2]. Finally, recent advancements in the 150–215 GHz range for the selective detection of semi-volatile explosive taggants are presented [3].

[1] F. Hindle et al., *Optica* 6(12), 1449-1454 (2019).

[2] C. Elmaleh et al., *Talanta* 253, 124097 (2023).

[3] M. Chrayteh et al., *Sensors and Actuators B: Chemical* 436, 137629 (2025).