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Direct-frequency-comb spectroscopy by a scanning Fabry-Pérot microcavity resonatorEdoardo Vicentini,^{1,2} Alessio Gambetta,^{1,2} Nicola Coluccelli,^{1,2} Yuchen Wang,^{2,3} Paolo Laporta,^{1,2} and Gianluca Galzerano^{2,*}¹*Dipartimento di Fisica, Politecnico di Milano, Piazza Leonardo da Vinci 32, 20133 Milano, Italy*²*Istituto di Fotonica e Nanotecnologie, Consiglio Nazionale delle Ricerche, Piazza Leonardo da Vinci 32, 20133 Milano, Italy*³*Department of Electronics and Nanoengineering and QTF Centre of Excellence, Aalto University, FI-00076 Aalto, Finland*

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We present a direct-frequency-comb spectroscopy method able to resolve the spectral comb structure with an instrumental frequency resolution limited only by the coherence properties of the comb source itself. The proposed technique, based on a 50 000-finesse scanning Fabry-Pérot microcavity resonator combined with a low-resolution diffraction grating, is characterized using a 250-MHz Er-fiber frequency comb demonstrating an ultimate frequency resolution as low as 50 kHz over an optical bandwidth up to 4 THz and with tens-of-milliseconds measurement times.

DOI: [10.1103/PhysRevA.102.033510](https://doi.org/10.1103/PhysRevA.102.033510)**I. INTRODUCTION**

Direct-frequency-comb spectroscopy (DFCS) is nowadays considered a fundamental tool in many areas of science and technology for high-fidelity mapping of atomic and molecular energy structure thanks to its unique performance: a combination of ultrabroad bandwidth, high-speed detection, high sensitivity, and absolute frequency accuracy [1–5]. Since the first DFCS experiments [6,7], several methods have been developed to increase the frequency resolution down to the comb mode level without sacrificing the broadband and fast acquisition time requirements [8–15].

Achieving high resolution is crucial especially for precision-spectroscopy applications where very narrow spectral features have to be detected over wide spectral bandwidths. In the last few years the number of experiments successfully exploiting DFCS for precision spectroscopy measurements has been rapidly increasing, with frequency resolutions generally ranging from tens of kilohertz to a few-gigahertz level, depending on optical bandwidth coverage and acquisition times. In the near-IR spectral region, for instance, by exploiting a pair of optical frequency combs (OFCs) tightly stabilized against narrow-linewidth cw optical references [16], dual-comb spectroscopy of Doppler-free two-photon transitions of 6-MHz linewidth has been performed, over a spectral span of 10 THz and with 1-MHz effective resolution (due to the frequency sampling interval) and 1-kHz theoretical resolution (limited by the comb linewidth). Single comb-tooth detection capabilities have also been demonstrated (across the same 10-THz optical bandwidth) by adaptive real-time dual comb spectroscopy [17], allowing for a theoretical resolution down to the ≈ 200 -kHz linewidth of the OFC source employed. Similar performance in terms of resolution has also been demonstrated by electro-optic (EO) dual-comb approaches, although over a more limited

optical bandwidth of ≈ 300 GHz. A remarkable result has also been achieved by Long and Reschovsky exploiting EO frequency-comb generation combined with a self-heterodyne detection scheme which allowed them to produce and detect optical frequency combs with spacing from 260 kHz down to 100 Hz, however over extremely narrow optical bandwidths, ranging, respectively, from 650 MHz to 40 kHz [18]. In the mid-IR spectral region, Fourier-transformed DFCS has been applied to measure buffer-gas cooled nitromethane absorption lines at ≈ 3 - μm wavelength with an effective resolution of ≈ 4 MHz (minimum theoretical resolution of 50 kHz due to the comb linewidth) over a single-scan bandwidth of ≈ 4 THz [19].

With few exceptions, among which is the work of Long and Reschovsky [18] where the system resolution has been verified by measuring electromagnetic-induced transparency as narrow as 1 kHz, the performance of the DFCS systems has, however, rarely been tested by actually measuring absorption features of linewidths comparable to the instrumental resolution. This is due to the difficulty of achieving the required extremely narrow optical features. Few groups have, however, made this effort, usually by locking a Fabry-Pérot (FP) cavity to the comb source and measuring its transmission features. For instance, the resolutions of Fourier-transformed and dual-comb spectroscopic approaches have been tested by measuring FP resonances of ≈ 200 -kHz and 2-MHz linewidths, respectively, over optical bandwidths wider than 15 THz [20,21]. Very recently, virtually-imaged-phase-array comb spectroscopy combined with a FP cavity to implement a Vernier scheme for filtering the comb mode line spacing demonstrated the capability to measure a 14-kHz-narrow FP resonance [22]. An alternative DFCS approach capable of resolving the mode structure of an OFC based on a scanning FP microcavity resonator, SMART (Scanning Micro-cavity Resonator), and a tunable single-frequency cw laser calibrator demonstrated resolution better than 20 MHz in 1-THz-wide optical bandwidth and 20-ms acquisition times, limited by the microcavity finesse and the adopted sequential

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FIG. 1. Experimental setup for the characterization of the SMART performance. AOM, acousto-optic modulator; BS, beam splitter; DAQ, data acquisition system; G, diffraction grating; GPS, global positioning system; M, mirror; PBS, polarizing beam splitter; PD, photodetector; PZT, piezoelectric transducer.

measurement scheme [14]. This last approach has the advantage of achieving performances comparable to the above systems but with a much more flexible architecture of lower complexity (being based on scanning FP cavity completely independent, not locked to the comb source adopted) and easily extendible to the important mid-IR region of molecular fingerprinting.

In this paper we present a significant advance in the SMART DFCS method exploiting parallel acquisition of two different microresonator transmission-mode orders. This allows us to improve the optical bandwidth for the single-scan acquisition while keeping the same accuracy level of the calibrated frequency axis and removing any tunability requirements for the cw laser. In this evolved implementation, calibration of the frequency axis is therefore achieved by exploiting a single-frequency cw laser with a fixed emission wavelength. Also, by means of a test FP cavity, we demonstrate the ability of the system to resolve spectral features as narrow as ≈ 300 kHz well below the 20-MHz linewidth of the SMART resonator transmission modes, ultimately limited by the comb source adopted. In particular, the full characterization of the proposed SMART DFCS method is performed at around $1.55 \mu\text{m}$ using a commercial self-referenced 250-MHz Er-fiber frequency-comb source, a free-running Er-fiber single-frequency laser, and a 50 000-finesse microresonator combined with a low-resolution dispersive grating.

Sub-MHz linewidth resonances of the test FP cavity as well as GHz linewidth acetylene absorption lines are employed as target spectroscopic spectral features in the Er-fiber comb wavelength region. Frequency resolution as low as 50 kHz in an optical bandwidth up to 4 THz and with few-tens-of-milliseconds single-scan measurement time is demonstrated, which compares with the best top-level DFCS implementations. The SMART technique being a direct measurement method, the signal-to-noise ratio (SNR) of the single-scan measurement is limited by the relative intensity noise of the comb source at a level of $\approx 1\%$ (for an integration bandwidth from 10 Hz to 2 MHz). By taking advantage of the coherent averaging of the recorded spectra, the SNR reaches the 10^3 level for a number of averages equal to 20 (corresponding to a measurement time of ≈ 1 s).

II. SMART DFCS EXPERIMENTAL SETUP

Broadband feasibility of the SMART method is demonstrated by precision spectroscopy of the $\nu_1 + \nu_3$ rovibrational band of acetylene (C_2H_2) at around $1.55 \mu\text{m}$, whereas the high-frequency resolution is validated by measuring the reflection coefficient of a sub-MHz linewidth performance-testing cavity. Figure 1 shows the implemented SMART setup based on a self-referenced dual-branch Er-fiber frequency comb with 250-MHz repetition

FIG. 2. (a) Microresonator transmission vs scanning time detected by PD₀ and PD₁. Several lines of the C₂H₂ P-branch absorption band are clearly visible as a modulation of the OFC spectrum detected by PD₁. Right panels: Temporal zooms around the cw laser, showing the microresonator frequency resolution (Δf) and the comb line spacing (f_r), and a detail of the resolved comb modes modulated by a C₂H₂ rovibrational absorption line. (b) Calibrated microresonator transmission detected by PD₀ and PD₁ vs absolute frequency. Bottom panels: Spectral zooms around the P8e and P6e C₂H₂ lines (red curves, HITRAN fitting profiles; bullets in red curves fit residuals).

rate, a narrow-linewidth distributed-feedback (DFB) Er-fiber laser, a scanning microresonator constituted by two spherical mirrors (1-m radius of curvature) with a finesse of $\approx 50\,000$ ($T = 0.005\%$ at 1540 nm) and a Free Spectral Range (FSR) of ≈ 600 GHz (corresponding to 250- μm mirror separation), and a reflective diffraction grating (600 grooves/mm). The microresonator transmission modes, the linewidths of which are ≈ 12 MHz, are swept by continuously scanning the mirror separation through a periodic voltage linear ramp applied to a piezoelectric transducer (PZT), as in the conventional optical spectrum analyzer configuration (corresponding to the case where only one comb mode is resonant with the cavity at a given time). To avoid spectral aliasing between the adjacent transmission orders of the FP microresonator, the microcavity transmission is first dispersed by means of a diffraction grating and then it is detected by two separate InGaAs photodetectors connected to a 12-bit digital oscilloscope: PD₀ measures the microresonator transmission-mode order, M , containing the cw laser frequency, whereas PD₁, mounted on a linear translational stage, detects higher transmission-mode orders, $M + K$, from $K = 1$ to 6 (enabling measurements spanning an optical bandwidth up to 4 THz). The main output of the OFC, providing 0.5-W average output power in the spectral region from 1500 to 1600 nm and with 250-MHz pulse repetition frequency, is combined with the DFB Er-fiber laser, emitting at 1550-nm wavelength, with a 10-ms linewidth of 10 kHz and ≈ 50 -MHz frequency accuracy. The combined radiation is sent to the performance-test cavity (1-GHz FSR with a finesse of ≈ 2500), back-reflected to the 10-cm-long C₂H₂ gas cell (filled at 50-mbars pressure), and finally coupled to the scanning microresonator for spectral analysis. The second output branch of the Er-fiber comb is used to detect the carrier envelope offset frequency, f_0 , by one-octave supercontinuum generation in a highly nonlinear fiber and an $f - 2f$ interferometer scheme. Self-referencing of the OFC is then obtained by stabilizing both the comb repetition and offset frequencies against a GPS-disciplined Rb radio-frequency standard (10^{-12} fractional frequency accuracy).

Figure 2(a) shows the single-scan spectra as acquired by the two photodetectors while scanning the microcavity length with a maximum span Δd of $\approx 0.5\,\mu\text{m}$ in 40 ms. The individual comb modes ($f_0 = 50$ MHz and $f_r = 249.986\,262\,7$ MHz) as well as the cw laser emission line are well resolved, with an instrumental linewidth [full width at half maximum (FWHM)] of ≈ 12 MHz, consistent with the 50 000-finesse value of the adopted microresonator. Absorption features of the C₂H₂ gas are clearly visible as a modulation of the comb spectrum detected by PD₁. By applying a calibration algorithm, the optical frequency of each comb mode is remapped onto an absolute frequency scale, allowing for the identification of the corresponding C₂H₂ absorption lines. The calibration employs the free-running cw laser, with a frequency stability much better than the comb spacing, as a frequency marker to measure the order, n_{cw} equal to the nearest integer of the ratio $\frac{(\nu_{\text{cw}} - f_0)}{f_r}$, of the comb tooth near the cw frequency. The frequencies of the comb modes are therefore uniquely determined using the comb equation $\nu_{\text{comb}} = [(n_{\text{cw}} + q)f_r + f_0]$, where the integer $q = \pm 1, \pm 2, \dots, \pm N$ for the M th-order microcavity transmission [see Fig. 2(a)] and $q = (K \frac{\text{FSR}}{f_r} \pm 1), (K \frac{\text{FSR}}{f_r} \pm 2), \dots, (K \frac{\text{FSR}}{f_r} \pm N)$ for the $(M + K)$ microcavity transmission order (where $K \frac{\text{FSR}}{f_r}$ is approximated to the nearest integer). In this way, absolute calibration of the frequency axis is obtained through the whole detected optical bandwidth in the same microcavity scanning time, allowing also for effective coherent averaging of the recorded spectra and corresponding increase of the SMART sensitivity [14]. Spectra detected by PD₀ and PD₁ are plotted in Fig. 2(b) versus absolute frequency, after calibration. By exploiting the spectral position of the cw reference ($\nu_{\text{cw}} = 193\,416.35 \pm 0.05$ GHz, $\lambda_{\text{cw}} = 1550.000$ nm), a value of 610.146 GHz can be retrieved for the microcavity FSR, corresponding to $M = 317$ and $M + K = 321$ ($K = 4$) for the transmission-mode order detected, respectively, by PD₀ and PD₁. Thanks to the absolute calibration of the frequency scale, limited only by the GPS-disciplined Rb radio-frequency standard, the assignment of the C₂H₂ rovibrational lines observed by PD₁ is therefore

FIG. 3. (a) Recorded transmission spectrum from a 10-cm-long gas cell filled with pure C_2H_2 at a pressure of 5 kPa (blue curve) over a bandwidth of ≈ 3 THz, corresponding to microresonator transmission-mode orders $K = 2, 3, 4, 5, 6$, together with the HITRAN database simulation for comparison (red curve). A mirror-image replica of the HITRAN spectrum (light gray) is also drawn underneath the recorded spectrum as a guide to the eye. (b) Retrieved line-center frequencies of the main C_2H_2 absorption lines.

performed with a line-center frequency accuracy better than 10 MHz. The limiting factors for the line-center frequency accuracy are mainly due to statistical contributions set by the finite SNR. In particular, for a single scan measurement a typical SNR of 200 is obtained as retrieved by the 0.5% root-mean-square (rms) value of the normalized residuals between the measured C_2H_2 transmission spectrum and the line fitting curves using a simple Voigt profile and the high-resolution transmission molecular absorption (HITRAN) database [23] [see Fig. 2(b), showing two spectral zooms around the $P8e$ and $P6e$ lines]. Taking into account the GHz-wide C_2H_2 absorption linewidth, this finite SNR translates into the observed 10-MHz uncertainty.

Sequential measurements of different microcavity transmission-mode orders have been performed by changing the position of the PD_1 photodetector to extend the overall optical bandwidth (without changing the microcavity FSR). Figure 3(a) shows the reconstructed C_2H_2 absorption spectrum together with a simulation based on the HITRAN database [23] over a bandwidth of ≈ 4 THz, corresponding to microresonator transmission-mode orders $K = 2, 3, 4, 5, 6$. Each measured spectrum is the average of 20 spectra recorded in a measurement time of 1.6 s, leading to a SNR of ≈ 1000 [14]. The retrieved center frequencies of the main C_2H_2 absorption lines are depicted in Fig. 3(b), showing a 2.2-MHz deviation with respect to the HITRAN database. In this averaged measurement, the uncertainty in the line-center frequency is mainly limited by a residual background modulation that slightly affects the position of the absorption-line-center frequencies (on the order of 1% of the measured linewidths, with respect to the HITRAN values).

In order to characterize the frequency resolution of the SMART technique we performed the detection of the periodic narrow-linewidth reflection profile of a test cavity. A plano-concave resonator configuration of 1-GHz FSR is chosen for this cavity so that, in resonance conditions, only one of four comb modes are transmitted through the cavity resonances. The high-reflectivity dielectric coatings of the resonator mirrors (fused silica substrates) have nominal reflectivities of 99.9 and 99.98% for the plane and concave (1-m radius of curvature) ones, respectively, in the wavelength range from 1520 to 1580 nm. The second surface of the curved mirror is antireflection coated whereas the input surface of the plane mirror is uncoated (Fresnel reflection coefficient of 3.3%). The test cavity finesse is measured at 1550 nm using the 10-kHz linewidth cw Er-fiber laser. A cavity linewidth (FWHM) of 380 kHz is measured by scanning the cw laser frequency across one of its resonances [see Fig. 4(c), bottom panel], corresponding to a cavity finesse of 2630, in very good agreement with the nominal mirror coating specifications and consistent with the measured cavity ring-down time of $0.84 \mu\text{s}$. To remove any frequency drift and environmental perturbations, one of the cavity resonances is frequency locked against the cw Er-fiber laser ($\nu_{\text{FP,cal}} = \nu_{\text{cw}}$) by means of wavelength modulation technique applying to the cw Er-fiber laser a frequency dithering (100-kHz modulation frequency and 100-kHz modulation depth) to its PZT actuator and closing the feedback control loop on the test cavity PZT. In addition, to fine tune the OFC around the test cavity resonances, a frequency-shifted replica of the OFC [obtained by using an acousto-optic modulator (AOM) frequency shifter] is stabilized with respect to the cw Er-fiber laser using the beat note signal between one tooth of OFC and the cw laser (beat note detection unit in Fig. 1).

FIG. 4. (a) Test cavity reflection profile measured by the DFCS SMART method in a 300-GHz optical frequency span. (b) Detail around the 193-THz frequency showing 27 cavity reflection profiles (top panel) and the retrieved resonance linewidths (bottom panel) as a function of optical frequency. (c) Comparison between the calibration reflection profile and linewidth measured by the DFCS SMART method (top panel) and by a cw Er-fiber laser (bottom panel). In all diagrams, the dotted red curves represent the computed values of the minimum reflectivity or resonance linewidths as a function of optical frequency due to the residual 3.3% reflectivity of the first surface of the test cavity input mirror whereas the full red curves are the cavity reflection profiles corresponding to a superposition of the real and imaginary parts of the complex Lorentzian profile [21].

This optical servo loop drives the fast intracavity electro-optic modulator to control the OFC repetition frequency in order to keep the following relation: $(\nu_{\text{comb}} + f_{\text{AOM}}) = (mf_r + f_0 + f_{\text{AOM}}) = \nu_{\text{cw}} + f_{\text{beat}}$. Also in this case, all the radio frequencies and the comb offset frequency are stabilized against the Rb clock. In this way, changing the beat frequency in the range from $f_{\text{AOM}} \pm \Delta f_{\text{AOM}}$, where $\Delta f_{\text{AOM}} = 1$ MHz, it is possible to finely scan the OFC under the calibration FP resonances and record the cavity reflection by the SMART technique. Figure 4(a) shows the FP cavity reflection in the spectral range from 192.86 to 193.16 THz as measured by the DFCS SMART method in a measurement time of 200 ms (average of 20 spectra acquired in 10-ms scan time). Periodical narrow peaks are clearly observable in the measured cavity reflection, with FSR equal to four times the comb repetition frequency and with a sinusoidal-like modulation of the minima in the range between 40 and 60%. This modulation is ascribed to the residual reflection of the first surface of the test cavity plane mirror, as pointed out by the excellent agreement between the experimental data and the theoretical dotted red curve corresponding to the simulated cavity reflection taking into account the 3.3% reflectivity of the uncoated mirror surface and the 6.35-mm thickness and 1.44 refractive index of the fused silica mirror substrate. This residual reflectivity turns out also in a slight modulation of the cavity finesse and of the resonance linewidths. Figure 4(b) shows 27 cavity reflection profiles and their retrieved resonance linewidths in the optical frequency around 193 THz. The cavity linewidths

(FWHM), obtained using a cavity reflection profile corresponding to a superposition of the real and imaginary parts of the complex Lorentzian profile [21], are well described by a sinusoidal function, $\Delta\nu_{\text{ref}} = \Delta\nu_0 + \Delta\sin[(2\pi\nu/c)2nt - \phi_0]$, with the following fitting parameters: $\Delta\nu_0 = 400 \pm 3$ kHz, $\Delta = 34 \pm 4$ kHz, $n = 1.44 \pm 0.05$ (mirror substrate refractive index), and $\phi_0 = 1.56\pi \pm 0.9$ rad, where the mirror substrate thickness is $t = 6.35$ mm and c is the speed of light. As a comparison, Fig. 4(c) reports the same cavity reflection profile measured using both the SMART DFCS method and the cw Er-fiber laser (by simply scanning the cw laser frequency across the auxiliary cavity resonance). It is worth pointing out the excellent agreement between the retrieved cavity linewidth values: 384 ± 19 kHz and 380 ± 3 kHz for the SMART DFCS and cw Er-fiber laser, respectively. In the case of comb measurements, the asymmetric cavity spectral shape, modeled by the superposition of the real and imaginary parts of the complex Lorentzian profile, is caused by a slight mismatch between the profile of the beam reflected at the cavity input mirror and that coming out by the multiple intracavity reflections. Indeed, the fine alignment of the test cavity was performed using the cw laser and some differences in coupling efficiency between the comb radiation and the calibration mode are due to chromatic aberration in the mode matching lenses. To better estimate the ultimate frequency resolution of the SMART method, we performed further line fittings using a complex Voigt profile instead of a complex Lorentzian profile to take into account any broadening contribution to the

test cavity linewidth due to instrumental limitation such as the comb tooth linewidth. The retrieved Gaussian (FWHM) linewidth contribution of the Voigt profile is 20 ± 44 kHz, i.e., consistent with zero and negligible with respect to the 384-kHz Lorentzian contribution reported in Fig. 4(c). In addition, the fit residuals of the complex Voigt profile are characterized by a rms value of 6% with respect to the resonance contrast, two times higher than the 3% value obtained with the complex Lorentzian profile shown in Fig. 4(c), indicating that the complex Lorentzian profile is more statistically appropriate than the Voigt one. However, in the worst case, assuming the deviation of the Gaussian linewidth as the instrumental contribution, the ultimate resolution of the implemented SMART DFCS method would be therefore at the 44-kHz level, which is consistent with the comb mode linewidth we previously measured in the range from 20 to 50 kHz for observation times from 1 to 10 ms [24].

III. CONCLUSIONS

We report on the full characterization of a compact and versatile direct-frequency-comb spectrometer with a resolving power higher than 4×10^9 in the spectral region

from 1.5 to $1.6 \mu\text{m}$, which is based on a high-finesse scanning Fabry-Pérot microresonator and a low-resolution diffraction grating combined with a free-running single-frequency cw calibration laser. The proposed method exploits the full potential of the optical frequency-comb source in terms of absolute frequency calibration, high sensitivity, and millisecond acquisition times, through parallel acquisition of two different microresonator transmission-mode orders (separated by several free-spectral-ranges) by using two independent photodetectors. The detection scheme lends itself to be extended to other spectral regions, in particular to the middle and far-infrared molecular fingerprint regions for precision spectroscopy and sensing applications.

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- [1] J. Mandon, G. Guelachvili, and N. Picqué, Fourier transform spectroscopy with a laser frequency comb, *Nat. Photon.* **3**, 99 (2009).
 - [2] A. Foltynowicz, P. Masłowski, T. Ban, F. Adler, K. Cossel, T. Briles, and J. Ye, Optical frequency comb spectroscopy, *Faraday Discuss.* **150**, 23 (2011).
 - [3] N. R. Newbury, Searching for applications with a fine-tooth comb, *Nat. Photon.* **5**, 186 (2011).
 - [4] N. Picqué and T. W. Hänsch, Frequency comb spectroscopy, *Nat. Photon.* **13**, 146 (2019).
 - [5] A. Marian, M. C. Stowe, J. R. Lawall, D. Felinto, and J. Ye, United time-frequency spectroscopy for dynamics and global structure, *Science* **306**, 2063 (2004).
 - [6] R. Teets, J. Eckstein, and T. W. Hänsch, Coherent Two-Photon Excitation by Multiple Light Pulses, *Phys. Rev. Lett.* **38**, 760 (1977).
 - [7] J. N. Eckstein, A. I. Ferguson, and T. W. Hänsch, High-Resolution Two-Photon Spectroscopy with Picosecond Light Pulses, *Phys. Rev. Lett.* **40**, 847 (1978).
 - [8] F. Keilmann, C. Gohle, and R. Holzwarth, Time-domain mid-infrared frequency-comb spectrometer, *Opt. Lett.* **29**, 1542 (2004).
 - [9] A. Marian, M. C. Stowe, D. Felinto, and J. Ye, Direct Frequency Comb Measurements of Absolute Optical Frequencies and Population Transfer Dynamics, *Phys. Rev. Lett.* **95**, 023001 (2005).
 - [10] S. A. Diddams, L. Hollberg, and V. Mbele, Molecular fingerprinting with the resolved modes of a femtosecond laser frequency comb, *Nature (London)* **445**, 627 (2007).
 - [11] C. Gohle, B. Stein, A. Schliesser, T. Udem, and T. W. Hänsch, Frequency Comb Vernier Spectroscopy for Broadband, High-Resolution, High-Sensitivity Absorption and Dispersion Spectra, *Phys. Rev. Lett.* **99**, 263902 (2007).
 - [12] A. Cingöz, D. C. Yost, T. K. Allison, A. Ruehl, M. E. Fermann, I. Hartl, and J. Ye, Direct frequency comb spectroscopy in the extreme ultraviolet, *Nature (London)* **482**, 68 (2012).
 - [13] G. Millot, S. Pitois, M. Yan, T. Hovhannisyan, A. Bendahmane, T. W. Hänsch, and N. Picqué, Frequency-agile dual-comb spectroscopy, *Nat. Photon.* **10**, 27 (2016).
 - [14] A. Gambetta, M. Cassinero, D. Gatti, P. Laporta, and G. Galzerano, Scanning micro-resonator direct-comb absolute spectroscopy, *Sci. Rep.* **6**, 35541 (2016).
 - [15] N. Coluccelli, M. Cassinero, B. Redding, H. Cao, P. Laporta, and G. Galzerano, The optical frequency comb fibre spectrometer, *Nat. Commun.* **7**, 12995 (2016).
 - [16] S. A. Meek, A. Hipke, G. Guelachvili, T. W. Hänsch, and N. Picqué, Doppler-free Fourier transform spectroscopy, *Opt. Lett.* **43**, 162 (2018).
 - [17] T. Ideguchi, A. Poisson, G. Guelachvili, N. Picqué, and T. W. Hänsch, Adaptive real-time dual-comb spectroscopy, *Nat. Commun.* **5**, 3375 (2014).
 - [18] D. Long and B. Reschovsky, Electro-optic frequency combs generated via direct digital synthesis applied to sub-Doppler spectroscopy, *OSA Continuum* **2**, 3576 (2019).
 - [19] B. Spaun, P. Changala, D. Patterson, B. J. Bjork, O. H. Heckl, J. M. Doyle, and J. Ye, Continuous probing of cold complex molecules with infrared frequency comb spectroscopy, *Nature (London)* **533**, 517 (2016).
 - [20] L. Rutkowski, A. C. Johansson, G. Zhao, T. Hausmaninger, A. Khodabakhsh, O. Axner, and A. Foltynowicz, Sensitive and broadband measurement of dispersion in a cavity using a Fourier transform spectrometer with kHz resolution, *Opt. Express* **25**, 21711 (2017).
 - [21] S. Okubo, Y.-D. Hsieh, H. Inaba, A. Onae, M. Hashimoto, and T. Yasui, Near-infrared broadband dual-frequency-comb

- spectroscopy with a resolution beyond the Fourier limit determined by the observation time window, [Opt. Express **23**, 33184 \(2015\)](#).
- [22] G. Kowzan, D. Charczun, A. Cygan, R. S. Trawicki, D. Lisak, and P. Masłowski, Broadband optical cavity mode measurements at Hz-level precision with a comb-based VIPA spectrometer, [Sci. Rep. **9**, 8206 \(2019\)](#).
- [23] I. Gordon, L. Rothman, C. Hill, R. Kochanov, Y. Tan, P. Bernath, M. Birk, V. Boudon, A. Campargue, K. Chance, B. Drouin, J.-M. Flaud, R. Gamache, J. Hodges, D. Jacquemart, V. Perevalov, A. Perrin, K. Shine, M.-A. Smith, J. Tennyson, G. Toon, H. Tran, V. Tyuterev, A. Barbe, A. Császár, V. Devi, T. Furtenbacher, J. Harrison, J.-M. Hartmann, A. Jolly, T. Johnson, T. Karman, I. Kleiner, A. Kyuberis, J. Loos, O. Lyulin, S. Massie, S. Mikhailenko, N. Moazzen-Ahmadi, H. Müller, O. Naumenko, A. Nikitin, O. Polyansky, M. Rey, M. Rotger, S. Sharpe, K. Sung, E. Starikova, S. Tashkun, J. V. Auwera, G. Wagner, J. Wilzewski, P. Wcisło, S. Yu, and E. Zak, The HITRAN2016 molecular spectroscopic database, [J. Quant. Spectrosc. Radiat. Transfer **203**, 3 \(2017\)](#).
- [24] N. Coluccelli, M. Cassinerio, A. Gambetta, P. Laporta, and G. Galzerano, Frequency-noise measurements of optical frequency combs by multiple fringe-side discriminator, [Sci. Rep. **5**, 16338 \(2015\)](#).