LASER PHYSICS

Dual-comb spectroscopy of water vapor with a free-running semiconductor disk laser

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Dual-comb spectroscopy offers the potential for high accuracy combined with fast data acquisition. Applications are often limited, however, by the complexity of optical comb systems. Here we present dual-comb spectroscopy of water vapor using a substantially simplified single-laser system. Very good spectroscopy measurements with fast sampling rates are achieved with a free-running dual-comb mode-locked semiconductor disk laser. The absolute stability of the optical comb modes is characterized both for free-running operation and with simple microwave stabilization. This approach drastically reduces the complexity for dual-comb spectroscopy. Band-gap engineering to tune the center wavelength from the ultraviolet to the mid-infrared could optimize frequency combs for specific gas targets, further enabling dual-comb spectroscopy for a wider range of industrial applications.

A fully stabilized optical frequency comb (OFC) consists of equally spaced frequencies with a relative uncertainty as low as 10^{-19} (*1*) and can serve as a very precise ruler for optical frequency measurements. The combs can be generated by mode-locked lasers with sufficient peak power in femtosecond pulses for supercontinuum generation in a single pass through a fiber or waveguide (*2*–*4*); generation with continuous-wave lasers (*5*) requires sufficient average power in a narrow-frequency linewidth to couple into a high-Q microresonator for highly nonlinear parametric interactions. Many applications require frequency combs in the 100-MHz to 10-GHz regime, where equally spaced microresonator combs become more challenging (*6*). Although mode-locked laser technology has enabled frequency-spacing stabilization since the 1980s (*7*, *8*), stabilizing the frequency comb offset requires substantial spectral broadening (*2*). To date, stabilized GHz OFCs have had to rely on rather bulky and complex ultrafast diode-pumped solid-state (*9*), fiber (*10*), or Ti:sapphire lasers (*11*). In comparison, optically pumped semiconductor disk lasers such as the mode-locked integrated external-cavity surface-emitting lasers (MIXSELs) (*12*) are often better suited for mass production and widespread applications, as they are based on a wafer-scale technology with reduced packaging requirements and potentially a higher level of integration.

Here we demonstrate dual-comb spectroscopy of water vapor with a single mode-locked laser cavity. The dual-comb mode-locked laser (*13*) generates two collinear, perpendicularly polarized OFCs with an adjustable frequency difference in comb spacing. A fast scan rate and the single–laser cavity approach support spectroscopic measurements even when using the free-running laser without any additional external spectral broadening or stabilization. This is a potential paradigm shift in frequency metrology: to use a narrowband OFC, which is then tuned to the desired spectral range on the basis of the specific target to be tested. This approach provides more power per comb line and a resulting higher signal-to-noise ratio (Fig. 1A). A dual-comb MIXSEL generates two OFCs from the same laser cavity and provides a potentially very attractive source for this approach. Tuning the OFC center wavelength from ultraviolet (UV) to mid-infrared (mid-IR) for different gas targets can be achieved with semiconductor band-gap engineering (*14*–*19*) (Fig. 1B). For the demonstration on water vapor, the dual-comb MIXSEL (Fig. 2A) was operated around a center wavelength of 968 nm with a pulse repetition rate of ≈1.7 GHz and a frequency offset of 4 MHz with an average power of >60 mW and a pulse duration of about 18 ps (fig. S1).

Dual-comb spectroscopy is generally very attractive for molecular spectroscopy (*20*, *21*), as well as asynchronous optical sampling (*22*), distance measurements (*23*), and fiber Bragg grating sensing (*24*). The technique often offers marked improvements in spectral resolution, sensitivity, and data-acquisition speed compared to traditional optical spectrometer techniques. With the dual-comb principle, a direct link between the optical frequencies and the microwave regime is established (Fig. 2B). This dual-comb approach is very attractive for applications such as spectroscopy. A key drawback of dual-comb spectroscopy is the requirement for two fully stabilized OFCs with slightly different comb spacing (Fig. 2B). For mode-locked lasers, this requires stabilization of both the pulse repetition frequency (i.e., the frequency comb spacing) and the carrier-envelope offset (CEO) frequency (i.e., the frequency comb zero offset) for two separate OFCs. Therefore, four separate stabilization loops and two highly nonlinear processes for supercontinuum generation are required.

There is a concerted ongoing effort in the research community to find ways to simplify these systems (*25*–*28*). The detection of the CEO frequency with optical *f*-to-2*f* interferometry (*2*) requires a coherent octave-spanning supercontinuum, which is very demanding on the laser performance and substantially reduces the power per mode. Unfortunately, most of the time the broad spectrum cannot be fully used for spectroscopy directly but needs to be filtered to avoid aliasing effects as described by the Nyquist-Shannon sampling condition (*29*). Therefore, very often it is sufficient to use a dual-comb source with a narrower optical spectrum centered at a wavelength that can be tailored to the region of interest (e.g., matching absorption lines of gases of interest).

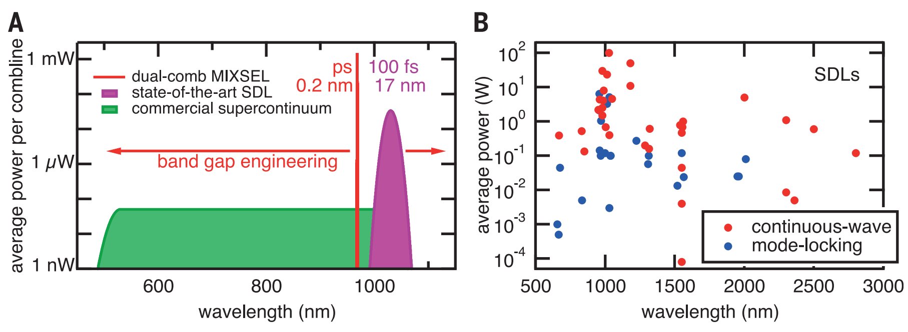

Fig. 1. Current state-of-the-art performance in frequency comb characteristics. (**A**) A commercial OFC (e.g., Menlo FC1500-250-WG MVIS) can cover an octave of optical spectrum. The large bandwidth, however, strongly reduces the average power per comb line to about 50 nW. Optically pumped semiconductor disk lasers (SDLs) can provide more than 1 mW of average power per comb line. With pulse durations as short as 100 fs, their optical spectrum spans up to 17 nm at a center wavelength around 1 μm (*41*). The center wavelength of ultrafast SDLs can be shifted with semiconductor band-gap engineering. (**B**) Large spectral coverage of optically pumped SDLs. Overview of continuous-wave and passively mode-locked SDL results in terms of center operation wavelength and average output power on a logarithmic scale. The plotted results are summarized in recent review papers (*14*–*19*).

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We consider the dual-comb MIXSEL an ideal source for dual-comb spectroscopy. Generally, given an optical source at the required wavelength, key requirements for molecular spectroscopy include (i) as much power per line as possible, resulting in better signal-to-noise ratio and faster measurement time; (ii) sufficiently narrow line spacing to resolve the spectroscopic feature (e.g., the mode-locked repetition rate equals the optical line spacing); and (iii) sufficiently narrow optical linewidth, of the individual comb lines, to resolve the spectroscopic features. For example, the water linewidth near 960 nm is ≈10 GHz; obtaining five sampling points per line requires a comb spacing of 1 to 2 GHz. Typical comb linewidth should be less than 10 to 100 MHz. MIXSEL combs have been successfully demonstrated in the 1- to 100-GHz regime (*30*), and in contrast to microcombs (*27*, *31*) or quantum cascade laser combs (*32*, *33*), the MIXSEL combs have shown even better performance in the 1- to 10-GHz regime so far. For example, without any further active stabilization, a single 2-GHz MIXSEL comb has a comb line–spacing variation of only $\approx 2.5 \times 10^{-4}$ integrated over a measurement time of 10 ms (*34*), which is a longer measurement time than required for most dual-comb spectroscopy applications (typically >1 μs to ideally ≈1 ms).

The simplicity of the dual-comb MIXSEL (Fig. 2A) allows for fundamental mode-locking in a simple straight linear cavity, and the operation wavelength can be adjusted with an intracavity etalon to a center wavelength of 968.3 nm (10,327 cm^{-1}) to match an absorption line of water vapor. The pulse repetition rate can be set by the laser-cavity length, which corresponds to about 9 cm in air for the 1.7-GHz comb spacing used in this work. For dual-comb mode-locked

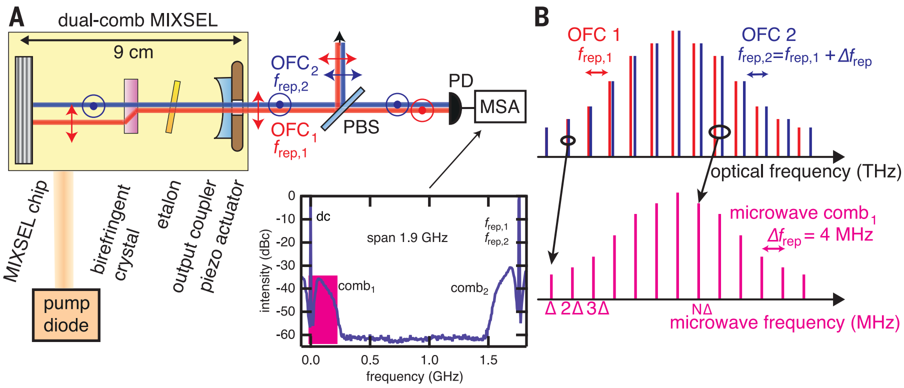

Fig. 2. Dual-comb source and operating principle. (**A**) Dual-comb MIXSEL. The diode-pumped dual-comb MIXSEL is a specialized SDL for which both the gain and the saturable absorber are integrated into the same semiconductor wafer. The semiconductor MIXSEL chip forms one cavity-end mirror, and the output coupler forms the other. In addition, two intracavity elements are used: first, an etalon to adjust the center wavelength, and second, a birefringent crystal for polarization splitting and dual-comb generation. The output coupler can be mounted on a piezo actuator to adjust the cavity length (and then the repetition rate). The dual-comb MIXSEL generates two collinear perpendicularly polarized (polarization indicated by circles and arrows) mode-locked pulse trains with a small difference in the pulse repetition frequency due to the difference in optical path length in the birefringent crystal. Thus, we obtain two collinear OFCs). A polarizing beam splitter (PBS), with an optical axis at an angle of 45° to the polarization axes of the OFCs, combines both OFCs into the same polarization, which then optically interfere on a photodetector (PD). A microwave spectrum analyzer (MSA) directly measures the microwave frequency comb, which manifests as a comb structure ($comb_1$) between dc and the pulse repetition rates $f_{rep,1}$ and $f_{rep,2}$. The pink rectangle indicates the microwave comb. dBc, decibels relative to the carrier. (**B**) The microwave frequency comb results from the optical interference on the PD between all the optical frequencies of the two collinear and combined OFCs with their offset in pulse repetition rate Δf_{rep}, which can be adjusted with the optical path length in the birefringent crystal. With this dual-comb principle, we obtain a direct link between the optical and the microwave frequencies.

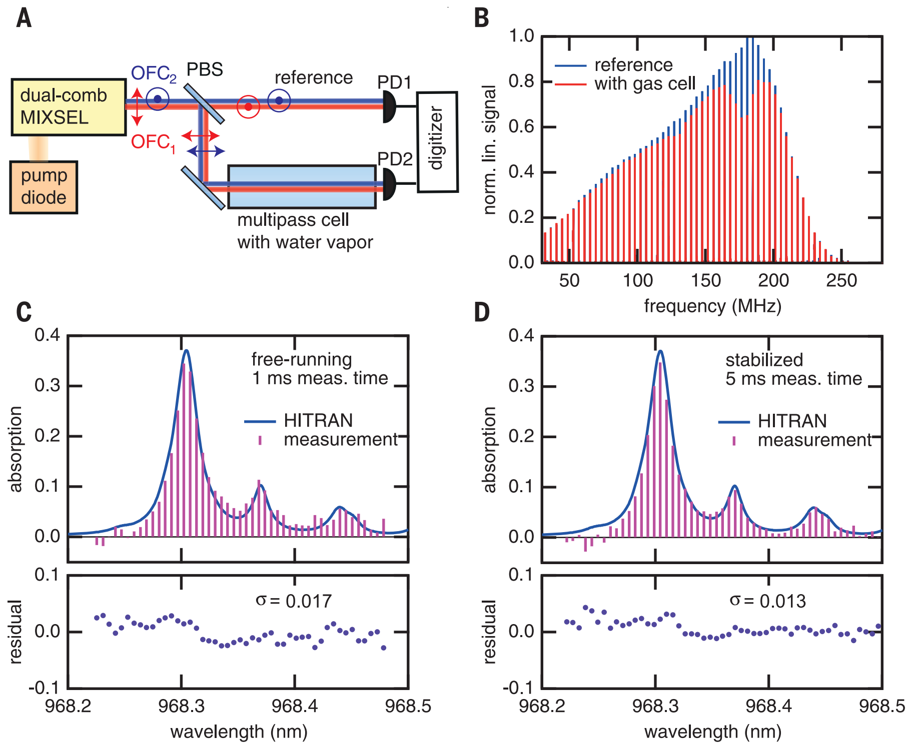

Fig. 3. Dual-comb spectroscopy results. (**A**) Schematic of the experimental setup with two combined dual-comb OFCs. A reference microwave spectrum is measured with PD1, and the microwave spectrum after the multipass gas cell is simultaneously measured with PD2, making the setup insensitive to phase errors. (**B**) Microwave frequency combs for the reference measurement (blue) and the measurement with the gas cell (red), which is modulated because of the absorption of the water vapor. (**C**) Absorption measurement with the free-running system on water vapor and comparison with the HITRAN database, with the residual error shown below in purple. (**D**) Same measurements as in (C) with the stabilized microwave comb.

operation (*13*), we insert a birefringent crystal inside this linear cavity, which splits the cavity beam on the side toward the MIXSEL chip into two spatially separated and cross-polarized beams with slightly different optical-cavity round-trip path lengths, defining a small difference in the comb frequency spacing Δf_{rep} of 4 MHz. This frequency difference can be adjusted by the thickness of the intracavity birefringent crystal (Fig. 2A). At the other side of the birefringent crystal, the two beams propagate collinearly but with perpendicular polarization, making them easy to separate, e.g., with a polarizing beam splitter (PBS). Therefore, both OFCs share the same optical components, pump laser, and electronics, leading to intrinsically strong mutual coherence.

When we first explored dual-comb modelocking (*13*), we encountered a very unexpected result. Cavity-length adjustments to stabilize one comb's spacing did not appreciably affect the other comb's spacing. We were concerned that the incoherent pump laser might be responsible, which would have been a more serious problem for dual-comb spectroscopy. We therefore had to first resolve this mysterious behavior. We gained insight into the underlying physics by comparing semiconductor and neodymium-doped:yttrium-aluminum-garnet (Nd:YAG) laser-gain materials, leading to the conclusion that the time delay introduced by the fully saturated saturable absorber is responsible for decoupling the two combs in an uncorrelated but deterministic way that can be fully stabilized (*35*). We therefore could then proceed to application of dual-comb spectroscopy.

Dual-comb spectroscopy of water vapor (Fig. 3) was first explored with the laser operating in free-running mode without any further active stabilization. A 40-m-long multipass cell was used to enhance the absorption, as the absorption cross section of water vapor at 968.3 nm is four orders of magnitude smaller than in the mid-IR. The two perpendicularly polarized collinear OFCs at the output of the dual-comb laser were combined into the same polarization with a PBS (Fig. 3A). One such combined dual-comb was directly detected with a photodetector (PD1) and a digitizer to serve as a reference (Fig. 3B, blue microwave spectrum). Analogous to Fourier spectrometers, the second frequency comb replaces the scanning mirror, which typically limits the scan rate to a few hertz in Michelson-type systems. With a frequency offset of 4 MHz between the two combs, we have a minimum acquisition time of only 0.25 μs (fig. S2) for the time-dependent interferograms detected by PD1. A simple Fourier transformation of the interferogram generates the spectrum shown in Fig. 3B. The other combined dual-comb after the PBS in Fig. 3A was sent through the air-filled multipass gas cell with a relative humidity of 60% at 36°C under ambient pressure and detected with PD2 (Fig. 3B, red microwave spectrum). The change introduced by the water-vapor absorption was clearly visible; the absorption was calculated by taking the difference between the two microwave spectra and normalizing the difference with the reference comb (Fig. 3B, blue microwave spectrum) (Fig. 3C) (*36*). Here we directly use the absorption line of water to calibrate the absolute wavelength (*36*), which is a commonly used method for dual-comb spectroscopy. A comparison with the high-resolution transmission molecular absorption (HITRAN) database (*37*) simulation using the laboratory parameters shows excellent agreement even though no active stabilization of the dual-comb laser was used. The residual error, with a standard deviation of 0.017, is shown below these data (Fig. 3C).

We could further improve the signal-to-noise ratio and prevent long-term drifts with a simple stabilization scheme. For example, a longer measurement duration of 5 ms with this active stabilization switched on shows even better agreement with the HITRAN data (Fig. 3D). For the stabilization scheme shown in Fig. 4A, both OFCs are superimposed in the same polarization on the photodetector (PD3) and analyzed with a microwave spectrum analyzer (MSA). The generated microwave frequency comb (Fig. 4B) results from the interference of the optical modes of the two beams, representing a direct link between the optical frequencies and the electronically accessible microwave regime (Fig. 2B). All lines of the microwave comb can be clearly resolved without any stabilization. With a measurement time of 10 s, the comb lines fluctuate by only ≈200 kHz,

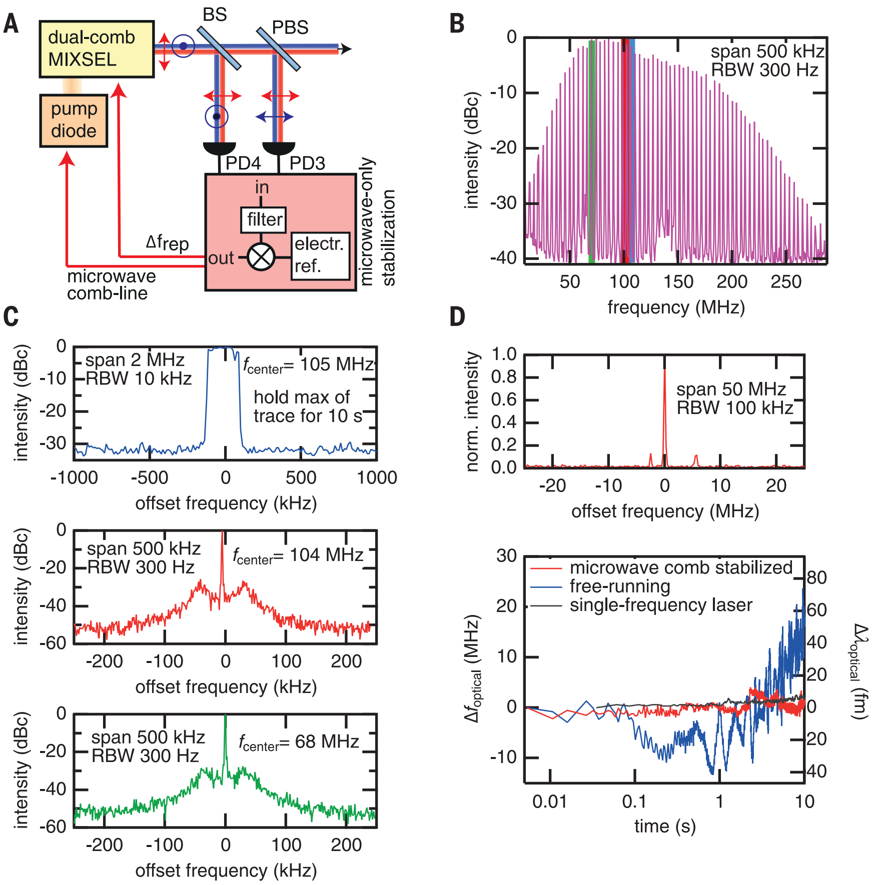

Fig. 4. Stabilization of the microwave comb and optical mode stability. (**A**) Schematic of the stabilization setup. By using a beam splitter (BS), both OFCs (red and blue lines) are superimposed in cross-polarization on PD4 to prevent optical interference and to simply measure and stabilize the difference in pulse repetition rate Δf_{rep}. With a PBS, both combined OFCs are superimposed in the same polarization on PD3, where the microwave comb is measured and one of the comb lines is stabilized. Electr. ref., electronic reference. (**B**) Microwave spectrum of the free-running microwave $comb_1$ (Fig. 2A) with a resolution bandwidth (RBW) of 10 kHz. (**C**) Magnification of a single microwave comb line. The blue line shows the fluctuations of a free-running comb line. The maximum of the trace is recorded for 10 s with a RBW of 10 kHz. The red line shows a stabilized comb line with a RBW of 300 Hz. The green line demonstrates that all comb lines are simultaneously stabilized if the red line, and additionally Δf_{rep}, are stabilized (RBW 300 Hz) (*36*). (**D**) Measurement of the absolute stability of the optical modes. The microwave spectrum shows the beat signal between the single-frequency laser and one optical comb line with a RBW of 100 kHz. The fluctuations of the beat frequency and the consequent fluctuations of the optical comb line are measured with a frequency counter over time in case of the free-running system (blue line) and with the microwave comb stabilization active (red line). The fluctuations of the single-frequency laser are measured with a wavelength meter (gray line). Stabilizing the microwave comb drastically improves the long-term stability of the optical comb lines.

shown here for a single comb line by holding the maximum of the trace for 10 s (blue line in Fig. 4C). This could be further improved with active stabilization. A single line (red line in Fig. 4B) of the comb was digitally filtered and locked to a stable electronic reference with an ultrahigh-frequency lock-in amplifier (UHFLI, Zurich Instruments). The error signal was corrected by current modulation to the multimode pump diode laser in the dual-comb MIXSEL. The stability of the comb line was substantially improved and the linewidth decreased to below 300 Hz (red line in Fig. 4C). In addition to one comb line, Δf_{rep} was stabilized by using a second input port of the UHFLI. The corresponding error signal was corrected by modulation of the cavity length by means of a piezo actuator–controlled output coupler. With both a stabilized comb line and a stabilized Δf_{rep}, all microwave comb lines were stabilized simultaneously, shown here for an arbitrarily chosen second comb line (green line in Fig. 4C) (*36*). Therefore, by applying only two feedback loops to signals that are measured directly with photodetectors without the need for any optical *f*-to-2*f*-interferometry (*2*), the full microwave frequency comb could be stabilized.

After characterizing the microwave frequency comb, we measured the absolute stability of the modes of the optical frequency combs. A single-frequency laser was used to generate a beat signal with one of the modes of the two optical OFCs (fig. S3). The strong beat frequency was measured at 100-kHz resolution bandwidth (RBW) with the MSA (Fig. 4D). The small side peaks originate from the beat between the single-frequency laser and a mode of the other optical spectrum, because the suppression is limited by the suppression ratio of the PBS. We then measured the beat frequency during 10 s every 5 ms with a gate time duration of 1 ms with a frequency counter to characterize the fluctuations of the optical mode (Fig. 4D). The measurement confirmed that the short-term (few milliseconds) stability of the optical modes is sufficient even for the free-running system. The long-term (few seconds) stability of the optical modes drastically improved by turning on the stabilization of the microwave comb. The measurement was not limited by the stability of the single-frequency laser, which was characterized with a wavelength meter (gray line in Fig. 4D). For a measurement time of 1 s, we could average more than 10^6 interferograms, and the standard deviation of the beat frequency in the case of the stabilized microwave comb was below 800 kHz (2.5 fm), which is more than three orders of magnitude smaller than the comb spacing of 1.76 GHz (5.5 pm). Because the linewidth of typical atmospheric absorption features is on the order of several gigahertz, an optical stability of 800 kHz is more than sufficient for most applications (*38*), and no further additional stabilization of the optical spectrum is required.

The main trade-off with the free-running dual-comb MIXSEL approach compared to current systems is the reduced frequency comb bandwidth, requiring different lasers for applications with substantially different center wavelengths. However, semiconductor technology allows the OFC center wavelength to be tailored to the region of interest through band-gap engineering from the UV to the mid-IR and can be very cost-effective because of its wafer scalability. We have observed a similar trend in biomedical imaging where the expensive, broadly tunable Ti:sapphire lasers are being replaced by much simpler optically pumped semiconductor disk lasers (SDLs) with a specific selection of operation wavelengths (*39*). In addition, we have shown that electrically pumped ultrafast SDLs are also possible with an average output power up to 50 mW to potentially further reduce complexity (*40*). We believe that the dual-comb MIXSEL approach has the potential to bring dual-comb spectroscopy from a laboratory environment to the field for a wide range of industrial applications.

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ACKNOWLEDGMENTS

We thank M. Kroner and A. Imamoglu for lending us the single-frequency laser (Toptica DL pro); J. Deiglmayr and F. Merkt for the wavelength meter (High-Finesse WS-7); and G. Villares, M. Rösch, and J. Faist for the frequency counter (Agilent 53220A). The authors acknowledge the support of the technology and cleanroom facility at Frontiers in Research: Space and Time (FIRST) of ETH Zurich for advanced micro- and nanotechnology. This work was financed by the Swiss Confederation program Nano-Tera.ch, which was scientifically evaluated by the Swiss National Science Foundation (SNSF). S.M.L., B. W. Tilma, M. Mangold, C. A. Zaugg, A. Klenner, and U.K. are inventors on a patent application (WO 2016/049787 A1) held and submitted by ETH Zurich that covers dual-comb mode-locking. Data can be obtained by contacting U.K. at keller@phys.ethz.ch.

SUPPLEMENTARY MATERIALS

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10 January 2017; accepted 1 May 2017
Published online 11 May 2017
10.1126/science.aam7424

Science

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Science **356** (6343), 1164-1168.
DOI: 10.1126/science.aam7424originally published online May 11, 2017

Two different combs from a single source

Combs of light divide the optical frequency spectrum into closely spaced tines that can measure molecular absorption spectra with exceptional precision. One appealing method to extend this precision down into the microwave regime is to simultaneously use two slightly distinct combs that differ in spacing by the magnitude of a microwave frequency. The challenge is ensuring that the combs remain synchronized. Link *et al.* solve this problem by generating both combs from the same semiconductor laser source. The resultant dual comb delivers highly accurate spectra of water vapor, and the approach could be generalized across the optical spectrum by tuning the semiconductor source.

Science, this issue p. 1164