Photo-acoustic spectroscopy (PAS) is one of the most sensitive non-destructive analysis techniques for gases, fluids and solids. It can operate background-free at any wavelength and is applicable to microscopic and even non-transparent samples. Extension of PAS to broadband wavelength coverage is a powerful tool, though challenging to implement without sacrifice of wavelength resolution and acquisition speed. Here, we show that the unmatched precision, speed and wavelength coverage of dual-frequency comb spectroscopy (DCS) can be combined with the advantages of photo-acoustic detection. Acoustic wave interferograms are generated in the sample by dual-comb absorption and detected by a microphone. As an example, weak gas absorption features are precisely and rapidly sampled; long-term coherent averaging further increases the sensitivity. This novel approach of photo-acoustic dual-frequency comb spectroscopy generates unprecedented opportunities for rapid and sensitive multi-species molecular analysis across all wavelengths of light.

In PAS\cite{1,2,3}, optical absorption of a modulated light source leads to periodic heating of a sample and the generation of an acoustic wave that can be detected by a microphone or an equivalent transducer (Figure 1a). As the detection relies on the acoustic waves (rather than a weak attenuation of an optical signal), photo-acoustic detection can be background-free, with high signal-to-noise ratio (SNR), and importantly, works at any wavelength of light. These unique properties have established PAS in environmental studies, solid state physics, chemical process control, medical application and life science, including for instance absorption measurements in atto-liter droplets\cite{4}, real-time monitoring of an ant’s respiration\cite{5} and in-vivo tomographic imaging\cite{6}. Quartz-enhanced photo-acoustic spectroscopy (QEPAS)\cite{7,8} and cantilever-enhanced photo-acoustic spectroscopy (CEPAS)\cite{9,10,11} have enabled ultra-sensitive trace gas detection below the part-per-trillion-level\cite{12,13}. Usually, PAS is performed at one single probing laser wavelength not ideal for the study of multiple species or studies in the presence of uncontrolled background absorption. Multiple laser sources can alleviate this problem to some extent, however, remain constraint to specific use cases. Therefore, in order to achieve broadband wavelength coverage, photo-acoustic detection has been combined with a Fourier-transform infrared spectrometer (FTIR-PAS)\cite{14}. The achievable resolution is determined by the scan range of the interferometer, which can reach several meters for high resolution instruments. In addition to incoherent light sources, such as super-continuum\cite{15}, coherent broadband spectra (unresolved optical frequency combs) have also been used to improve the overall performance\cite{16,17}, and might allow adopting techniques for sub-nominal resolution\cite{18} in the future. As such FTIR-PAS represents a powerful tool for broadband photo-acoustic spectroscopy, how-

![Photo-acoustic dual-frequency comb spectroscopy](image-url)
ever, its resolution and acquisition speed remain limited due to the mechanical scan of the interferometer.

In this work, we show that the unmatched precision, speed and wavelength coverage of dual-frequency comb spectroscopy (DCS) can be combined with the advantages of photo-acoustic detection. This novel approach of photo-acoustic dual-frequency comb spectroscopy (PA-DCS) enables the rapid and scan-free acquisition of absorption features with high resolution and precision via photo-acoustic dual-comb multi-heterodyne detection.

Figure 1 illustrates the concept of PA-DCS. Similar to conventional DCS, two frequency combs are used in our demonstration whose optical frequency components \( \nu_n^{(i)} \) are described by

\[
\nu_n^{(i)} = n \cdot f_{\text{rep}}^{(i)} + \nu_0^{(i)},
\]

where \( f_{\text{rep}}^{(i)} \) and \( \nu_0^{(i)} \) denote the repetition rate (i.e. the comb line spacing) and the combs’ optical offset frequencies, respectively. The index \( i = 1, 2 \) distinguishes the two combs, and \( n = 0, \pm 1, \pm 2, \ldots \) are the comb line indices. The combs’ repetition rates and offsets differ only by small amounts \( \Delta f_{\text{rep}} = \left| f_{\text{rep}}^{(1)} - f_{\text{rep}}^{(2)} \right| \ll f_{\text{rep}}^{(1,2)}, \) and \( \Delta \nu_0 = \left| \nu_0^{(1)} - \nu_0^{(2)} \right| \ll f_{\text{rep}}^{(1,2)}, \) so that pairs of optical comb lines \( \nu_n^{(1)} \) and \( \nu_n^{(2)} \) are only separated by acoustic frequencies. When both combs are optically combined, this can be interpreted as a single frequency comb

\[
\tilde{\nu}_n = n \cdot \left( f_{\text{rep}}^{(1)} + f_{\text{rep}}^{(2)} \right) + \frac{1}{2} \left( \nu_0^{(1)} + \nu_0^{(2)} \right),
\]

whose optical lines are modulated in optical power according to \( 1 + \cos (2\pi f_n t) \) with frequencies

\[
f_n = \left| \nu_0^{(1)} - \nu_0^{(2)} \right| = n \cdot \Delta f_{\text{rep}} + \Delta \nu_0.
\]

Exposing the sample to the dual combs, it experiences periodic heating with frequency \( f_n \) if light at the optical frequency \( \tilde{\nu}_n \) is absorbed. The periodic heating will lead to the generation of heterodyne acoustic waves in function of the absorbed power. The superposition of all acoustic waves constitutes a \( \Delta f_{\text{rep}} \)-periodic interferogram that is detectable by a microphone or an equivalent transducer. In order to be detectable, all acoustic frequencies \( f_n \) need to respect the bandwidth limitation of the transducer.

Key to our demonstration are dual-frequency combs with high mutual coherence that enable dense packing of the acoustic multi-heterodyne beatnotes \( f_n \) within the microphone’s bandwidth. Dual combs with high mutual coherence have been implemented in various ways based on mode-locked lasers or electro-optic modulation, and have also been extended to the infrared molecular fingerprint regime. In this proof-of-concept demonstration, we use two near-infrared electro-optic combs (Figure 2). However, the concept can be readily applied to the mid-infrared domain. In order to ensure high mutual coherence between both electro-optic combs, they are derived from a single continuous-wave (CW) laser with an optical frequency of \( \nu_{\text{CW}} = 195.09 \) THz (1536.71 nm). The amplified CW laser is split into two beams, each traversing first an acousto-optic modulator (AOM) where the laser frequencies are shifted by \( f_0^{(1)} = 80 \) MHz and \( f_0^{(2)} = 80 \) MHz + 4 kHz, respectively (i.e. \( \nu_0^{(1,2)} = \nu_{\text{CW}} + f_0^{(1,2)} \)), to create a relative comb offset of \( \nu_0 = 4 \) kHz. Next, each beam passes through an electro-optic modulation (EOM) stage that includes one intensity and two phase modulators to generate a series of approximately 40 comb lines, spaced by \( f_{\text{rep}}^{(1)} = 1 \) GHz and \( f_{\text{rep}}^{(2)} = f_{\text{rep}}^{(1)} + 125 \) Hz, respectively. In total, both combs deliver 20 mW of average power for photo-acoustic detection. A simple aluminum tube (diameter 4 mm, length 10 mm) whose ends are sealed by two angled glass windows serves as the photo-acoustic sample chamber. Attached to the sidewall of the tube and connected through a small hole is an off-the-shelf micro-electro-mechanical system (MEMS) microphone with 20 kHz bandwidth. A battery-powered amplifier and digitizer is used to record the acoustic signals for a memory limited duration of up to 1 hour. The repetition rate difference of \( \Delta f_{\text{rep}} = 125 \) Hz was chosen so that all acoustic multi-heterodyne beatnotes would be within 4 to 6 kHz and well within the microphone’s bandwidth. Given the combs’ high mutual coherence, we note that more beatnotes (i.e. more optical sampling points) could readily be accommodated, particularly, when lowering \( \Delta f_{\text{rep}} \) or selecting a transducer with a larger bandwidth. The combs are sent through the sample tube in a multi-pass configuration. A small fraction of the comb light is sent to a photo-detector that provides a reference for normalization of the photo-acoustic signal and also enables an increase of the combs’ mutual coherence in post-processing. In a wavelength regime where suitable photo-detectors may not be available, it could be replaced by photo-acoustic detection of black-body absorption. Both the microphone as well as the photo-detector signals are sampled. All modulation sources are synchronized to a 10 MHz frequency standard to ensure precise sampling and coherence in the acquisition process.

As a spectroscopic target we choose acetylene gas (C2H2) at atmospheric pressure and lab temperature as it provides precisely defined and interference-free absorption features. In a first experiment, the absorption cell is filled with acetylene gas, giving rise to the heterodyne acoustic interferogram signal shown in Figure 2. Figure 2 shows two examples of the heterodyne acoustic spectra after Fourier-transformation (PA-DCS signal) for acquisitions with a duration of 80 ms (10 interferograms) and 800 ms (100 interferograms), respectively. As expected, a longer acquisition time yields a higher SNR in the PA-DCS signal. In contrast to conventional DCS, photo-acoustic multi-heterodyne beatnotes are only gen-
FIG. 2. Experimental setup and results. **a,** Experimental setup for the photo-acoustic detection of gaseous acetylene (C$_2$H$_2$). A tunable CW laser is amplified by an erbium-doped fiber amplifier (EDFA) and used as a common seed for the generation of two optical frequency combs via acousto-optic and electro-optic modulation (AOM, EOM). (COL: free space collimator; RD: reference (photo-)detector MIC: low-noise MEMS microphone; see main text for more details). **b,** Acoustic multi-heterodyne signal recorded by the microphone (5 interferograms; after high-pass filtering) for an acetylene filled cell. **c,** Spectrum of the acoustic multi-heterodyne signal (acquisition time 80 ms and 800 ms). Inset: Multi-heterodyne reference spectrum as recorded by the reference (photo-)detector (over the same span). **d,** Acetylene absorption signature obtained after normalizing the acoustic multi-heterodyne spectrum by the reference spectrum and comparison to the HITRAN model.

In order to retrieve the true absorption profile, the acoustic multi-heterodyne beatnotes are normalized to account for the uneven spectral power envelope of the combs. Here, we accomplish this by dividing the PA-DCS signal (Figure 2c) by the photo-detected heterodyne reference beatnotes (inset in Figure 2c). The mapping of the acoustic to the optical frequency axis is described by Eqs. 2 and 3 (Note: this implies a scaling factor of $(f^{(1)}_{\text{rep}} + f^{(2)}_{\text{rep}})/(2\Delta f_{\text{rep}}) \approx 8 \times 10^6$ between acoustic and optical frequency axes). The resulting C$_2$H$_2$-absorption signature is shown and compared to the HITRAN-based prediction in Figure 2. Blue dots show the absorption retrieved from an 800 ms acquisition and shaded areas (grey, yellow, blue) represent the standard-error intervals for different durations of acquisition (8 ms, 80 ms and 800 ms). Excellent agreement between the HITRAN-prediction (red line) and the measured absorption profile is achieved, with the smallest residuals (below 3% relative to peak absorption) observed with an 800 ms acquisition. The grey shaded area demonstrates that a fast, 8 ms acquisition (i.e. a single interferogram) is sufficient to retrieve the coarse features of the absorption profile. The spectral resolution for each sampling point is given by the combs’ absolute optical linewidth (here: ~100 kHz), so that instrumental lineshape effects are negligible (resolution 5 orders of magnitude below the width of the absorption feature). Moreover, the frequency spacing of the sampling points (1.0000000675 GHz) is precisely defined by the mean repetition rate of the two combs (Eq. 3).

Next, we investigate the extent to which even longer recordings of time $\tau$ can increase the SNR. To explore this, the cell is filled with N$_2$-diluted C$_2$H$_2$ with a concentration of 1% and probed by combs centered at 1532.83 nm. Acquisitions of different duration are processed (similar to what is shown in Figure 2c) and the SNR of the highest acoustic beatnote (at 4 kHz) is determined as a function of $\tau$. Indeed, as Figure 3 shows, the SNR increases with $\tau$ (yellow trace), however, it markedly deviates from the $\tau^{1/2}$-scaling one would expect in the scenario with perfect noise-averaging. This deviation is due to small and slow length fluctuations in the non-common optical path of the combs that limits their mutual coherence on the time scale of few seconds or longer. These slow fluctuations manifest themselves
FIG. 3. **Long-term acquisition.** Signal-to-noise ratio (SNR) with (blue) and without (yellow) phase correction as a function of acquisition time $\tau$. The black line indicates the ideal case where the SNR increases proportionally to $\tau^{1/2}$. Inset: Spectrum of an heterodyne acoustic beatnote for $\tau = 1000$ s with (blue) and without (yellow) phase correction.

As phase drifts in the multi-heterodyne beatnotes, which fortunately, can easily be tracked and corrected for numerically$^{25,43,45}$ Here, we extract the phase drift (one phase value for all heterodyne beats) from the reference heterodyne signal and, after low-pass filtering ($<0.1$ Hz), subtract it from the phase of the heterodyne acoustic beatnotes. This *a-posteriori* phase-correction extends the effective mutual-coherence time of the combs by compensating for the slow path length fluctuations. As shown by the blue trace in Figure 3, phase correction results in an increase of the SNR close to the ideal scaling (black line) up to the maximal recording duration of 1 hour. This result suggests that even longer recordings could be leveraged to further increase the signal to noise ratio. A small deviation from the ideal scaling is observed for acquisitions longer than 300 s and attributed to residual differential phase drifts between the heterodyne beatnotes, which could be addressed by tracking the phase of each beatnote separately. To further illustrate the effect of phase correction, the inset in Figure 3 shows the spectrum of an individual heterodyne acoustic beatnote for a recording time of 1000 s. With phase correction applied (blue trace), a narrow 1 mHz linewidth heterodyne beatnote is detected. Without phase correction (yellow trace) the drifting beatnote has a reduced SNR. Generally, in photo-acoustic gas spectroscopy, the SNR depends on the used optical power, the absorption coefficient, the probing wavelength, the photo-acoustic cell design$^{[50]}$, the microphone, the surrounding matter, environmental conditions (pressure, temperature) as well as the recording duration. In the current proof-of-concept configuration, based on the SNR in Figure 3, we estimate a minimal detectable noise equivalent $C_2H_2$ concentration of 10 ppm for a recording time of 1000 s.

In conclusion, we have demonstrated photo-acoustic dual-frequency comb spectroscopy (PA-DCS) as a novel spectroscopic technique that can achieve high resolution, rapid acquisition and sensitive detection. Owing to its principle, PA-DCS can operate at any wavelength and across the large spectral bandwidth accessible to frequency combs$^{[51,52]}$. In the mid-infrared and other wavelength regimes where photo-detection is challenging, it can complement alternative DCS approaches, such as those based on optical field sampling or up-conversion$^{[51,52]}$. In future work, PA-DCS could be combined with high power quantum-cascade dual-laser comb$^{[53,54]}$, cavity enhancement$^{[55]}$, multi-MHz bandwidth optical microphones$^{[56]}$, as well as opto-mechanical transducers$^{[57,58]}$ to further increase sensitivity, spectral coverage and acquisition speed. As such, our demonstration generates new opportunities for rapid, sensitive broadband multi-species molecular analysis across all wavelengths of light.

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