Incoherent broadband cavity enhanced absorption spectroscopy using supercontinuum and superluminescent diode sources

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Abstract: We investigate incoherent broadband cavity enhanced absorption spectroscopy using a tailored supercontinuum source. By tailoring the supercontinuum spectrum to match the high reflectivity bandwidth of the mirrors, we achieve an unprecedented spectral brightness of more than 7 dBm/nm at wavelengths where the effective absorption path length in the cavity exceeds 40 km. We demonstrate the potential of the source in spectrally broadband measurement of weak overtone transitions of carbon dioxide and methane in the near-infrared 1590 nm – 1700 nm range and evaluate its performance against that of a typical superluminescent diode source. Minimum detectable absorption coefficients (3σ) of $2.2 \times 10^{-9}$ cm$^{-1}$ and $6.2 \times 10^{-9}$ cm$^{-1}$ are obtained with the supercontinuum and the superluminescent diode sources, respectively. We further develop a spectral fitting method based on differential optical absorption spectroscopy to fully and properly account for the combined effect of absorption line saturation and limited spectral resolution of the detection. The method allows to cope with high dynamic range of absorption features typical of real-world multi-component measurements.

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1. Introduction

Cavity ring-down spectroscopy (CRDS) [1], integrated cavity output spectroscopy [2], NICE-OHMS [3], broadband CRDS [4], and cavity-enhanced dual-comb spectroscopy [5] are all spectroscopy techniques which allow for significant increase in sensitivity as compared to direct absorption measurement due to the long optical path in the cavity. These techniques generally require the use of a narrowband continuous wave laser which limits their applicability to single species measurements, or of an optical parametric oscillator system for wavelength scanning, or of a mode-locked laser and a cavity mode matching scheme which increases the complexity and costs.

Incoherent broadband cavity-enhanced absorption spectroscopy (IBB-CEAS) [6] on the other hand offers similar sensitivity and allows for selective multi-component measurement due to the broad bandwidth of the light source. It is simple and robust, which makes it ideal for portable setups, and indeed in situ measurements of NOx and HONO have been recently demonstrated using this technique [7]. One drawback, however, associated with IBB-CEAS is the resulting low total transmittance of the cavity due to the incoherent nature of the light source. This limits the measurement sensitivity by (i) decreasing the signal-to-noise ratio, and (ii) by limiting the maximum value of the mirrors’ reflectivity that can be used. Yet, compared to coherent CW methods where the ring-down signal is obtained after momentarily locking the laser source to a cavity mode, IBB-CEAS possesses a higher duty cycle which partly compensates for this disadvantage.

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Fiber-based supercontinuum (SC) sources exhibit unique characteristics including very high brightness, broad bandwidth and quasi-perfect spatial coherence, which makes them attractive candidates for spectroscopic applications [8,9]. Recently, SC sources have been employed in IBB-CEAS measurement setups [10–13]. Yet, the bandwidth of the sources has been filtered significantly to match that of the mirrors' spectral reflectivity, resulting in total intensity of only a few milliwatts. Superluminescent diodes (SLDs) have also been used, as they are more cost-effective and may possess similar brightness [14]. In this work, we demonstrate the potential of cost-effective SC sources tailored for IBB-CEAS in the near-IR. Specifically, we use a compact source with extremely high spectral brightness over the whole high reflectivity bandwidth of the cavity mirrors, which allows us to take full advantage of the IBB-CEAS benefits. The source is built using a cost-effective nanosecond fiber laser pump and a standard single-mode fiber. Compared to previous studies, our SC source has about two orders of magnitude higher spectral brightness. In order to illustrate the advantages of our source over other traditional light sources, we perform a systematic comparison of the SC source performance vs. that of a SLD where the SLD was selected to best match the IBB-CEAS setup.

We further develop a convenient differential optical absorption spectroscopy (DOAS) method for the data analysis that correctly accounts for the saturation of absorption lines in a low-resolution measurement. In contrast to previous approaches, our method is able to cope with a wide range of strong and weak absorption features typical of real-world multi-component measurements, leading to a greatly enhanced detection dynamic range. This customized DOAS-method is then used in both, calibration of the instrument as well as the retrieval of trace gas concentrations from the measurement samples. Combined with our DOAS method, we demonstrate that our SC source results in a significantly enhanced sensitivity and bandwidth compared to the SLD.

2. Experimental setup

To generate the supercontinuum, we inject 700-ps-pulses from an ultra-compact fiber laser (Keopsys Kult) operating at 1.5 μm into the anomalous dispersion regime of a standard single-mode fiber (Ocean UltraWave SLA). The result is a broadband supercontinuum with more than 250 nm total bandwidth extending from 1500 nm to beyond 1750 nm (the limit of our spectrum analyzer) with a total output power of 800 mW. Figure 1(a) shows the output spectrum of the SC source for different values of input pulses peak power. In the anomalous dispersion and long pump pulse regime, SC generation is dominated by noise-seeded modulation instability and soliton dynamics [15]. These processes are clearly evident in Fig. 1(a); as the pump peak power is increased, the spectral intensity of the symmetrical sidebands around the pump resulting from modulation instability increases. In the time domain, this corresponds to a random modulation that develops on top of the pulse envelope. This process eventually breaks the long input pulse into a random train of breathers that evolve into short solitons as a result of higher-order dispersion and stimulated Raman scattering [16]. The solitons experience the soliton self-frequency shift, resulting in efficient energy transfer from the pump towards the long wavelengths. We note that although the spectrum of the SC generated in this way fluctuates heavily from pulse to pulse, the average spectrum is remarkably stable when using a high repetition rate laser. The sharp peak at the pump wavelength corresponds to the remaining background of the pulse envelope in the time domain (mostly at the leading and trailing edges where the intensity is too low for breathers to emerge). This spectral peak is typical for a SC generated with long pump pulses and may problems in specific applications. In particular, the coherent laser peak may momentarily lock to the IBB-CEAS cavity modes through optical feedback, causing high intensity transmission through the cavity at the laser pump wavelength and resulting in significant stray light noise in the spectrometer.

The SLD source on the other hand produces 17 mW of average power with a spectral bandwidth (FWHM) of 43 nm. A direct comparison between the SLD and SC spectra [see Fig. 1(b)] clearly illustrates the advantage of using the SC source for broadband spectroscopy.
with a power-bandwidth product more than 300 times that of the SLD. The SC spectrum also matches the overall spectral shape of the mirrors’ reflectivity but does not significantly exceed their usable bandwidth. This contrasts with previous approaches using SC sources where an extremely broadband source was filtered to match the mirrors’ reflectance spectrum and using only a tiny fraction of the available optical power.

Fig. 1. (a) Supercontinuum spectrum as a function of pulse peak power. (b) Comparison of supercontinuum (SC) and superluminescend diode (SLD) spectra together with the mirror reflectance curve. The reflectance is calculated from the transmittance curve measured by the manufacturer assuming zero losses.

The experimental IBB-CEAS setup is illustrated in Fig. 2. Light from the SC is first collimated using a fiber collimator (D = 3.0 mm) and then slightly focused into a 1.17 meters long near-confocal cavity containing the trace gas sample using a plano-convex lens (f = 750 mm). The mirrors (Layertec 102103) have a radius of curvature of 1 m with a reflectivity exceeding 99.99% between 1520 nm and 1670 nm. Such high reflectivity results in an effective path length exceeding 10 km, but the average transmission of the cavity is less than $0.5 \times 10^{-4}$, i.e. half of the transmission of a single mirror [17]. The maximum intensity of the SC spectrum corresponds to the maximum of the mirrors’ reflectivity, thus corresponding to the wavelength of minimum transmission. This is particularly important as it allows to significantly increase the signal-to-noise ratio. Light exiting the output mirror of the cavity is coupled into a multi-mode fiber and the absorption spectrum is recorded using an optical spectrum analyzer (Ando AQ6315B) with a 0.1 nm resolution and integration time of 0.5 s, resulting in a scanning speed of c.a. 10 nm/min.

The cavity is airtight with valves for gas input and output, and the construction allows for easy angular adjustment of the mirrors. A visible He-Ne-laser is used for initial alignment of the cavity. We then switch to the SC or SLD source and adjust the mirrors’ alignment to maximize the total transmission. The slightly non-confocal geometry of the cavity results in a quasi-continuous transverse mode-structure. To further average out the mode structure, and to prevent mode-locking effects resulting from optical feedback into the laser, the output cavity mirror is vibrated mechanically at 620 Hz. In the case of the SC source, a slight misalignment of the cavity is used to suppress any remaining mode-locking. All of the results presented here were measured using a small constant gas flow. The cavity was purged using N$_2$ flow between measurements.
3. DOAS analysis of IBB-CEAS data

Broadband measurement has many benefits beyond the obvious potential for multi-component analysis. However, retrieving accurately concentrations from broadband spectral data is a nontrivial task. Arguably, the most advanced method of broadband trace gas analysis is differential optical absorption spectroscopy (DOAS), originally developed for long-range atmospheric measurements [18]. In DOAS, a modeled differential absorption spectrum is fitted to the experimental spectrum using a least squares method. The concentrations are then retrieved as the fitting parameters. Because of its ability to reliably retrieve concentrations even in the presence of overlapping background absorbers, this method can be very useful in the measurement of large molecules such as many volatile organic compounds which have broad and smooth absorption spectra. When measuring smaller molecules, an important benefit is the ability to separate the absorption of molecules (which varies rapidly as a function of wavelength) from the smoothly varying background signal caused by e.g. aerosol extinction and system drifts. This can be achieved by high pass filtering the spectrum or by including a low-order polynomial in the fit. In order to use a DOAS based fitting method in IBB-CEAS, a model is required for cavity enhancement of broadband spectra. The model should account for the wavelength-dependent mirror spectral reflectivity, possible system drifts, and the instrument resolution.

The theory of cavity-enhancement for incoherent sources is well known. Originally derived in the time-domain [6], the same result can be obtained using a more intuitive frequency-domain approach [13]. Yet, the results derived in [6,17], when used in the context of IBB-CEAS [10–14] require modification for proper implementation of the limited spectrometer resolution. The results in [6,17] are given for the absorption coefficient and this has generally led authors to implement the resolution by convolving the absorption cross section with the instrument slit function, which is unphysical because the resolution affects the measured transmittance spectrum and not the properties of the sample. Accounting for the spectrometer resolution in this way can then lead to significant distortions in the measured spectrum of gas with strong and narrow absorption features that saturate in the cavity, since the saturation effect is now underestimated in the model. On the other hand, modeling accurately the limited spectrometer resolution is especially important because (i) saturation occurs in a regime where the cavity enhancement of the SNR is largest, (i.e. where the single pass absorption losses are comparable to mirror losses [19]), and (ii) the spectral resolution is typically a limiting factor when building a compact and cost-effective instrument making best use of the simplicity and robustness of IBB-CEAS technique. When the spectral resolution is properly accounted for in the model, the dynamic range of the detection can be greatly increased due to the fact that the weaker absorption features still provide information when the stronger lines are saturated. This will also reduce the error caused by strong saturated background absorbers such as water vapor. Similar complications affecting broadband CRDS spectra have been discussed in detail previously, together with a method for proper analysis of the affected spectra [4]. Here, we develop a specific model for IBB-CEAS that correctly...
accounts for the absorption line saturation and limited spectrometer resolution. The model can be used with a least squares method fitting procedure to retrieve concentrations (or other parameters) from measured spectra using DOAS principles.

The saturation of absorption lines is explicitly discussed in the context of IBB-CEAS in [20]. We start from the intermediate result given in Eq. (26) in [20] that considers the integrated “cavity enhanced differential transmittance”:

\[
\frac{I(\lambda)}{I_0(\lambda)} = \frac{\rho(\lambda)}{\rho(\lambda) + \tau(\lambda)},
\]

where \(I(\lambda)\) and \(I_0(\lambda)\) are the output intensities in the presence and absence of absorbing species, respectively. The term \(\rho(\lambda)\) represents the mirror losses (dominated by the mirror transmission, assuming high quality mirrors). Using this notation, the mirror reflectance is given by \(R(\lambda) = 1 - \rho(\lambda)\). The wavelength-dependent term \(\tau(\lambda)\) is the small single-pass gas absorption given by the linear approximation of Beer-Lambert law, where transmittance of the sample gas can be written as \(T(\lambda) = 1 - \tau(\lambda)\), and

\[
\tau(\lambda) = \sum_j \sigma(\lambda)_j N_j d_0,
\]

where \(\sigma(\lambda)_j\) are the absorption cross sections of the gas species, \(N_j\) their number densities and \(d_0\) is the cavity length. We model the measured transmittance in the presence of gas sample by convolving the cavity transmittance with an instrument slit function \(g(\lambda)\). It is important to take the instrument resolution into account at this step, since it directly affects the transmittance spectrum. The optical density is then given as the inverse of the natural logarithm of the transmittance. Finally, we add a low degree polynomial (3rd degree typically is enough) to take into account background effects such as smoothly varying absorbers, aerosol extinction and system drifts. We arrive at the following equation for modeled optical density:

\[
D_{\text{fit}}(\lambda) = -\ln\left(\frac{\rho(\lambda)}{\rho(\lambda) + \sum_j \sigma(\lambda)_j N_j d_0} \cdot g(\lambda)\right) + \sum_k a_k \lambda^k,
\]

where the number densities \(N_j\) and polynomial coefficients \(a_k\) are used as fitting parameters. The absorption cross sections \(\sigma(\lambda)_j\) can be modeled beforehand using a spectral database, e.g. HITRAN 2008 [21], and well-known line broadening mechanisms.

4. Instrument calibration

In order to calibrate the instrument and retrieve concentrations from DOAS fits, mirror losses \(\rho(\lambda)\) need to be characterized. Using a reference sample is the most common approach but a ring-down measurement using a diode laser [22] and broadband phase-shift CRDS [23] have also been employed. It is important to note that the total losses not only depend on the properties of the mirrors, but also on the coupling parameters and alignment of the cavity. Therefore, using a separate source such as in [22] may not allow an accurate determination of the mirror losses. Here, we therefore use a reference sample to calibrate the instrument. More specifically, we measure the optical density spectrum from a sample of known concentration, then use a Levenberg-Marquardt algorithm to fit Eq. (3) to the measured spectrum with \(\rho(\lambda)\) containing the fitting parameter. As a starting point for \(\rho(\lambda)\), we use the mirror transmittance provided by manufacturer and add a small constant fitting parameter independent of wavelength to represent additional losses such that \(\rho(\lambda) = \rho(\lambda)_{\text{trans}} + \epsilon\). This correction parameter \(\epsilon\) is then the retrieved from the least squares fit. We note that a more accurate broadband calibration is also possible with this method, since the constant \(\epsilon\) can be replaced with a curve \(\epsilon(\lambda)\), for example a low degree polynomial, whose coefficients then act as the calibration parameters. Doing so could in principle increase the accuracy of the calibration at wavelengths away from the center wavelength, provided the reference sample has absorption
features covering the whole high reflectivity bandwidth of the mirrors. This would result in a smoothly-varying calibration curve for the mirror losses as function of wavelength, similarly to what can be obtained using a point-by-point calibration approach [23].

Due to the different cavity alignment in each case, characterization of the mirror losses was performed separately for both the SC and SLD light sources using a reference sample of 100 ppm CO$_2$ and whose weak 3v$_1$ + v$_3$ combination overtone transitions are at the center of the mirror bandwidth in the 1595 nm – 1620 nm range. The absorption cross-section $\sigma(\lambda)$ was modeled using the HITRAN 2008 database [21] and by applying the appropriate Voigt broadening of the lines at the measurement temperature and pressure. By fitting the modeled optical density spectrum to the experimentally recorded spectrum, we obtained a mirror loss parameter $\epsilon = 12.1 \times 10^{-6}$ for the SC source and $\epsilon = 8.8 \times 10^{-6}$ for the SLD source, respectively. The discrepancy arises from the fact that when employing the SC source, the cavity was intentionally slightly misaligned to prevent mode-locking effects resulting from optical feedback. The corresponding maximum effective path lengths were also calculated for reference. The maximum effective path length is solely determined by the average lifetime of the photons in the cavity and depends on the mirror losses and cavity length [6]. Using a value $\rho(\lambda_0)_{\text{trans}} = 16 \times 10^{-6}$ for the minimum center bandwidth transmission losses (measured by the manufacturer) and the correction parameters $\epsilon$ given above, we estimate the maximum effective path lengths to be 40 km and 46 km for the SC and SLD source, respectively.

5. Results and discussion

The performance of the calibrated instrument with the SC and SLD sources was evaluated by measuring the concentration of CO$_2$ and CH$_4$ samples whose weak 3v$_1$ + v$_3$ combination overtone and 2v$_3$ overtone transitions, respectively, fall within the measurement bandwidth of the setup. We first measured the absorption spectrum of CO$_2$ with 200 ppm concentration. Figure 3 shows the experimental optical density spectrum measured using the SC source (red solid line). The result of the least squares fit using Eq. (3) is superimposed (blue solid line), in excellent agreement with the measured spectrum. A concentration of 194 ppm was retrieved from the fit. The experiment was subsequently repeated using the SLD source. The results (not shown here) are very similar to those obtained with the SC source except for a slightly noisier spectrum. In the case of the SLD source, a concentration of 196 ppm was retrieved from the fit. We note that in this regime of relatively high optical density and limited spectrometer resolution, applying the usual methodology [10–14] of implementing the spectrometer resolution in the model resulted in significant distortion of the modeled spectrum, causing a large error in the retrieved concentration.

In order to evaluate the performance in lower SNR conditions, we subsequently characterized a CO$_2$ sample with a lower concentration of 40 ppm. Figure 4 shows the experimentally recorded absorption spectra measured with the SC source [Fig. 4(a)] and the SLD source [Fig. 4(b)]. We can see how in this case the significantly lower brightness of the

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Fig. 3. (a) Experimental CO$_2$ absorption spectrum measured using the SC source from a sample containing 200 ppm of CO$_2$ (red) together with the modeled least squares fit (blue). Concentration of 194 ppm was retrieved from the fit. (b) Close-up of the fit, highlighting the excellent agreement between measured and modeled spectra.

Fig. 4. (a) Experimental CO$_2$ absorption spectrum measured using the SC source from a sample containing 40 ppm of CO$_2$ (red) together with the modeled least squares fit (blue). Concentration of 194 ppm was retrieved from the fit. (b) Close-up of the fit, highlighting the excellent agreement between measured and modeled spectra.
SLD source results in a much more noisy spectrum. There is also a noticeable baseline shift in both spectra, caused by system drifts including light source intensity and cavity coupling variations that can occur in-between and during the recording of the I and \( I_0 \) signals. Broadband DOAS spectral fitting methods are, however, rather effective in retrieving concentrations even in cases where the spectrum appears noisy and shows baseline shifts. By fitting Eq. (3) to the spectra of Fig. 4, concentrations of 38 ppm and 45 ppm were retrieved for the SC and SLD measurements, respectively. The error is larger in the SLD case as can be expected from the excess of noise. Nevertheless, in both cases, including the compensation for the baseline drift that was accounted for by the low-degree polynomial in Eq. (3), the modeled spectra showed good agreement with the measured spectra.

![Fig. 4. Experimental CO\(_2\) absorption spectrum measured from concentration of 40 ppm using (a) supercontinuum (SC) and (b) superluminescent diode (SLD) source. Concentrations of (a) 38 ppm and (b) 45 ppm were retrieved using DOAS fits.](image)

We then evaluated the detection limit of the instrument for CO\(_2\). For this purpose, we repeatedly measured the CO\(_2\) concentration from the cavity when flushed with dry nitrogen, such that no CO\(_2\) was actually present. For both light sources the background spectrum was scanned 66 times and 66 \( I/I_0 \) pairs were formed from the consecutive signals. We note that determining the detection limit by scanning \( I_0 \) and I spectra immediately one after another does not take into account the time needed to fill the cavity with the gas sample. However, we believe that this is not a major issue, since instrument drifts are effectively compensated by the polynomial in Eq. (3). Although the optical density spectra calculated from the 66 pairs showed no sign of trace gas absorption, the CO\(_2\) absorption spectrum was nevertheless fitted to the measured spectra. Histograms of retrieved concentrations showed Gaussian distributions around zero concentration with standard deviations of 0.41 ppm for the SC source and 1.12 ppm for the SLD source. From these values, we can define the 3\( \sigma \) detection limits for CO\(_2\) as 1.2 ppm and 3.4 ppm for the SC and SLD source, respectively, with a scanning time of 2.5 minutes. The factor of three difference in the detection limits is somewhat surprising given that the SC has roughly ten times more spectral brightness at the CO\(_2\) measurement bandwidth. This would be the expected result if the detection was shot-noise limited, but because we are measuring weak signals that are also affected by the thermal noise of the detector, one might expect a larger benefit from the increased brightness. We attribute this discrepancy mainly to the complications caused by the slight misalignment of the cavity to decrease the unwanted effects of optical feedback. This intentional misalignment caused a 13% reduction in the effective path length as well as a reduction in the effective transmission of the cavity, which contribute to decreasing the sensitivity enhancement in the SC case. There is also possibly small amplitude noise in the SC spectrum caused by residual...
optical feedback. The misalignment could be avoided by using an optical isolator or a spectral filter to suppress the optical feedback at the pump laser wavelength, which could improve the sensitivity. Finally, we note that evaluating the measurement stability for both sources by means of Allan variance analysis is an important task, best performed using a faster detection scheme, such as e.g. a detector array, and it is left here as a subject of future work. Here, the scanning speed of 10 nm/min was approximated to be a good compromise between minimizing detection noise and system drifts.

The detection limits can be generalized by calculating the minimum detectable absorption coefficients for equivalent single-wavelength measurements done using the strongest line in the spectrum. In the $3v_1 + v_3$ band, the strongest line has an absorption cross section of $7.56 \times 10^{-23}$ cm$^2$. By using this value for absorption cross section and by assuming that detection limits of 0.41 ppm and 1.22 ppm were achieved in hypothetical single-wavelength measurements using this line only, we calculate the minimum detectable absorption coefficients (3σ) of $2.2 \times 10^{-9}$ cm$^{-1}$ for the SC source, and $6.2 \times 10^{-9}$ cm$^{-1}$ for the SLD with a measurement time of 2.5 minutes. For convenience, the optical parameters and performance of both sources are summarized in Table 1.

Table 1. Optical parameters and performance of SC and SLD sources

|                         | Supercontinuum | Superluminescent diode |
|-------------------------|----------------|------------------------|
| Center wavelength (nm)  | 1598           | 1604                   |
| -3dB bandwidth (nm)     | 142            | 55                     |
| Average power (mW)      | 800            | 17                     |
| Max. spectral brightness (dBm/nm) | 7.1            | -4.7                   |
| Maximum path length (km) | 40             | 46                     |
| CO$_2$ detection limit (ppm) | 1.2            | 3.4                    |
| MDA$^c$ ($10^{-9}$ cm$^{-1}$) | 2.2            | 6.2                    |

Ignoring the peaks around 1545 nm in SC spectrum. $^c$Minimum detectable absorption coefficient.

The sensitivity of our measurements is comparable to that reported in [10] and [13]. The fact that the achieved sensitivity with our SC source is similar might be surprising at first sight, given that our SC source has roughly two orders of magnitude higher spectral brightness. However, these previous studies used detection schemes with a silicon CCD and an FTIR spectrometer, respectively, which allow light to collect much more efficiently than the scanning optical spectrum analyzer employed here. Yet, we anticipate that with a CCD or FTIR, our approach would allow for a significantly better sensitivity.

Besides providing a better sensitivity in the measurement due to its higher spectral brightness, the SC source has the additional benefit of a much broader bandwidth. We demonstrated the broadband potential of the SC source by measuring the absorption spectrum of methane that is over 80 nm wide and covers a large fraction of the mirrors’ high reflectivity ($R > 99.995\%$) bandwidth. This could not be performed with the SLD source due to its limited bandwidth (see Fig. 1). The results for a sample with a nominal concentration of 4 ppm are shown in Fig. 5. The DOAS fit (not shown) using the same procedure as with CO$_2$ is in good agreement with the measurements and produces a concentration value of 3.2 ppm. The value differs by 20% from the nominal concentration, which is more than what could be expected. We attribute this discrepancy to two factors: the nonlinearity of the optical scanning spectrometer at wavelength ranges near the edge of its operational bandwidth at 1750 nm and the fact that the mirror loss calibration was performed using only a wavelength-independent correction parameter near the center of their spectral bandwidth. The latter issue could be addressed by using higher-order correction parameters as discussed in Section 3.
The IBB-CEAS setup presented here could be improved upon in a variety of ways. Firstly, replacing the scanning spectrometer with a detector array could greatly increase the SNR and thus the sensitivity through a higher duty cycle. This would also decrease the cost of the setup and make it more portable. In this case, the lower resolution would make the data analysis methods presented here even more crucial. Secondly, optimizing the cavity and coupling parameters according to guidelines given in [19] and suppressing optical feedback through the use of an optical isolator or spectral filters could increase the optical throughput and SNR even more. Using higher-order correction parameters in the mirror loss calibration would also improve the accuracy of the instrument. Finally, the measurement bandwidth could be further increased by using a prism cavity [11].

6. Conclusions

In conclusion, we have demonstrated incoherent broadband cavity-enhanced absorption spectroscopy using a tailored fiber-based supercontinuum source and very high reflectivity mirrors in the near-IR. Upon systematic comparison with a superluminescent diode source, the supercontinuum was shown to offer better sensitivity and much broader usable bandwidth. The minimum detectable absorption coefficients (3σ) were $2.2 \times 10^{-9}$ cm$^{-1}$ for the SC source and $6.2 \times 10^{-9}$ cm$^{-1}$ for the SLD source with a scanning time of 2.5 minutes. For both sources, the sensitivity is higher than what has been typically demonstrated for broadband CRDS [4] and does not fall greatly behind the most advanced single-wavelength CRDS methods where the minimum detectable absorption coefficients are of the order of $10^{-11}$ cm$^{-1} – 10^{-10}$ cm$^{-1}$ [24–27]. Compared to single-wavelength methods, IBB-CEAS has better selectivity, flexibility, and true potential for multi-component analysis. We have also developed and demonstrated a DOAS-type method for data analysis that that correctly accounts for the saturation of absorption lines in a low-resolution measurement, which is crucial for real-world high dynamic range multi-component detection. Finally, we note that extending the technique to mid-infrared wavelengths offers exciting possibilities for ultra-sensitive multi-component measurements in the molecular functional group region, where strong absorption bands associated with fundamental vibrational transitions can be used.

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Fig. 5. Experimental broadband CH$_4$ absorption spectrum measured from a concentration of 4 ppm using a supercontinuum source. Concentration of 3.2 ppm was retrieved from the DOAS fit.