CW INTEGRATED CAVITY OUTPUT SPECTROSCOPY

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Abstract

A new approach is described that enables continuous, narrow band laser sources to be used in conjunction with the recently developed integrated cavity output spectroscopy (ICOS) technique to obtain sensitive, quantitative absorption spectra in a simple experimental configuration. Absorption data obtained with CW-ICOS are related to the classical Fabry-Perot intracavity absorption model, which describes why the intracavity absorption is enhanced. In this study, a method of continuously injecting cw laser light into the cavity is described, as is a simple means of interpreting the ICOS data to extract accurate absorption intensities. Absorption spectra of vibrational combination bands of CO_2 and H_2O in the 1.3 μm region are presented as a proof-of-principle of the approach.

Introduction

Recently, the integrated cavity output spectroscopic technique (ICOS) was introduced and demonstrated using pulsed dye laser sources [1]. In the ICOS approach, light is coupled into a highly reflective optical cavity, and the cavity output is measured in order to extract intracavity losses such as molecular or atomic absorption. The initial ICOS work demonstrated that the technique could be employed to obtain absorption spectra with high sensitivity, comparable to that obtained with other sensitive techniques such as cavity ringdown spectroscopy. In ICOS, the extinction coefficient for intracavity samples is obtained by measuring the total transmitted output of the cavity as the input light source is scanned in frequency. Per pass extinction coefficients are derived through knowledge of the cavity mirror reflectivity at the injection laser wavelength. The initial, proof-of-principle ICOS studies were performed with a pulsed dye laser, where normalization of the integrated output was achieved by measuring the intensity of the input light source, which needed to measured at approximately 1% accuracy to infer a per pass fractional absorption of $1\text{x}10^{-6}$. This work demonstrated that very weak absorptions could easily be quantitatively measured using a pulsed dye laser with 10-15 % shot-to-shot amplitude fluctuations.

In this letter, a variation of the ICOS technique, cw-ICOS, is described that enables absorption spectra to be obtained using continuous sources such as diode lasers. This cw version of the ICOS method employs two strategies to effectively eliminate the problems traditionally associated with the frequency selectivity or sharp resonances of the optical cavity. Under coherent excitation, cw pumped optical cavities exhibit high-energy buildup when the cavity mode coincides with the excitation wavelength, resulting in near-unity "transmission" when on-resonance and near zero transmission when off-resonance. The resulting contrast of the cavity is a function of the cavity stability as well as the degree of mode matching achieved between the laser and the cavity. Small fluctuations in the cavity can result in large deviations in transmitted energy, making traditional absorption measurements impossible. A high finesse optical cavity also acts as an efficient spectral filter to narrow band light passed through it, which can result in strong variations in transmitted energy with source frequency.

In cw-ICOS, both of these potential problems are eliminated by effectively randomizing the cavity mode structure on a time scale significantly faster than the frequency-scanning rate of the injection light source. This is achieved by dithering both the frequency space of the optical cavity as well as that of the input light source. The modulation of the source laser frequency eliminates transmission fringes with variation in wavelength. In the present study, we demonstrate that when the depth and frequency of the cavity and laser source modulation is sufficient, the ICOS method of integrating the total cavity output can be employed to obtain accurate, linear absorption spectra. Here absorption spectra are obtained in the 1.3 micron region using a frequency scanned, narrow bandwidth diode laser source. These studies establish the feasibility of the concepts employed in cw-ICOS, and demonstrate the utility, generality, and simplicity of the technique.

Theory

In ICOS, the absorption signal is obtained through the integration of the total signal transmitted through a ringdown-type, optical cavity absorption cell, in much the same fashion as in conventional absorption measurements. Single pass cavity losses are calculated from the measured cavity output, which is a function of the mirror reflectivity as well as scattering and absorption losses, which occur due to the presence of samples located between the mirrors. In a fashion similar to that employed in Cavity Ringdown measurements [2], molecular or atomic absorption is determined by measuring the baseline transmission of the cavity, and normalizing the transmitted signal to this value. This removes the effects of spectrally broad effects such as Rayleigh scatter from the measurement. The enhancement of the ICOS absorption signal results from the effectively infinite sample path length, as the light retraces the same path on each cycle. In a manner different from that used in either conventional multipass or ringdown-based absorption measurements, the ICOS measurement represents an *asymptotic* transmission value, due to this essentially infinite sample path length. The measured absorption changes to the integrated output are thus very large, even in the case of weak (per pass) absorption. The ICOS signal for a pulsed cavity injection has been shown [1] to be:

$$I = I_0 T^2 e^{(-kl)} \times [2\log(R')]^{-1} \qquad (1)$$

where I is the transmitted intensity, I_O is the incident light intensity, T is the mirror transmission, k is the intra-cavity absorption per unit length, L is the cavity sample length, and R′ is an effective "reflectivity". The effective reflectivity embodies the effect of intracavity absorption on the Q of the cavity, and is given by

$$R' = Re^{(-kl)} \qquad (2)$$

where R is the “true” mirror reflectivity. The change in the ICOS signal is very nearly linear when the magnitude of the absorption is small relative to the total mirror transmission losses (ignoring mirror scatter).

The predicted ICOS sensitivity and dynamic range have been demonstrated for pulsed injection into a sample cavity [1]. Here, we examine what occurs when coherent, continuous optical injection of the cavity is applied. In cases where the cavity is actively stabilized, the cavity behaves like an ideal classical Fabry-Perot etalon. The transmission characteristics of such cavities are well established and the effects of factors such as absorption are easily calculated [3,4]. The "enhancement" of intra-cavity absorption in Fabry-Perot type optical cavities a well known phenomena, and results in a stronger peak absorption value at the center of the cavity mode. In this case, the steady state cavity transmission for the center of the mode is given by

$$I/Io = \{1 - [A/(1-R + A)]\}^2 \times \boldsymbol{A} \qquad (3)$$

where A is the fractional absorption per optical pass (or reflection), T is the fractional transmission per pass in the cavity, and ***A*** is the Airy function. Here, it is important to note that this value represents the theoretical value for the absorption enhancement on line center for a stabilized optical cavity. This "enhancement" does not suggest that the actual molecular absorption strength is modified. Rather it is a direct result of the increased optical residence time of light within a highly reflective cavity.

In the present work, we demonstrate a method of reproducibly injecting optical energy into a cavity with amplitude stability such that small changes in measured transmission (or integrated output) can be usefully related to intra-cavity absorption. In this approach, continuous laser light is injected into a linear, two-mirror cavity, which is aligned, such that the light retraces the same optical path with each pass, as is the case in Cavity Ringdown cavities. When the laser frequency is scanned over several free spectral ranges, the expected cavity transmission spectrum is the classic Fabry-Perot modulated pattern. When the laser beam and cavity mirrors are properly aligned, the observed transmission pattern clearly shows sharp longitudinal cavity mode structure. Typically, low order transverse modes are also excited, though to a much lesser extent. There are two approaches to achieving a stable and reproducible transmission as the injection laser frequency is scanned. One approach would be to employ stabilized optical cavities and laser sources, locked to each other, and simultaneously scanned such that the injection laser frequency always matches a cavity mode. Such an approach is very sensitive to vibrations, and moreover results in high intracavity energy buildup, which can lead to undesirable saturation effects. In the present approach, we spoil the finesse of the cavity of the cavity such that the period of time in which the laser and cavity are in coincidence is not sufficient for the theoretical energy buildup to be achieved. This is achieved by rapidly modulating the cavity length while simultaneously modulating the input laser frequency, resulting in a rapid randomization of the cavity and laser dynamic mode matches. When averaged over a number of these dynamic mode matches, the time average transmission is found to be nearly constant as the laser is scanned in injection frequency. The cavity length is modulated using a piezoelectric actuator as one of the cavity mirror mounts, in a fashion similar to that employed in the cw-Cavity Ringdown studies [5,6], while the laser is frequency modulated, in this case using a piezoelectric actuated grating mechanism in an external cavity diode laser. The experiments described below demonstrate that when the modulation frequency and depth are properly adjusted, this strategy enables cw ICOS

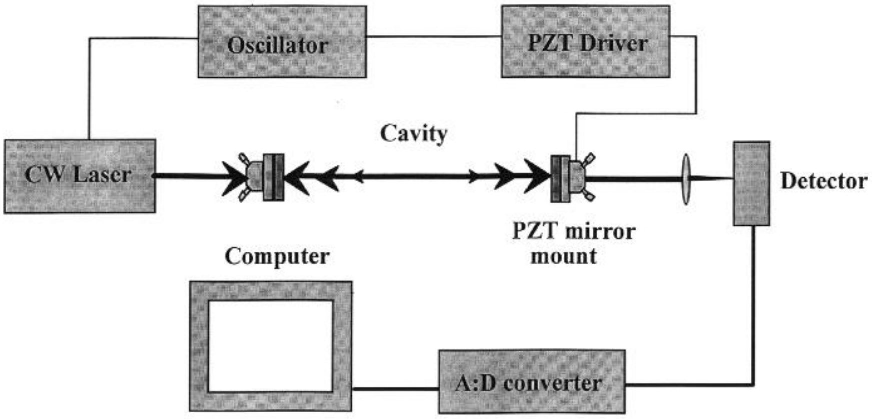

Figure 1. Schematic representation of the cw-ICOS experimental configuration. A single mode output diode laser operating near 7550 cm^{-1} was coupled to a two mirror cavity comprised of mirrors with reflectivities of 0.9994, and the averaged transmitted signal detected using a InGaAs detector interfaced to a LabView computer analysis system.

measurements to be performed to yield quantitative, cavity enhanced absorption spectra.

Experimental

The cw ICOS experimental apparatus is shown in Fig. 1, which also displays an oscilloscope that was used for diagnostic purposes only. Absorption measurements were made using a two mirror optical cavity (R=99.94% @1.3 μm) employing a single mode, tunable diode laser (New Focus model 6224) as the injection light source. The system was tuned continuously between 7630 cm^{-1} and 7410 cm^{-1}. Absorption measurements were made in air for several cavity sizes, and in a closed cell that could be evacuated and filled with controlled pressures of various gases.

The cavities ranged from 30 cm to 50 cm in length, resulting in cavity free spectral ranges of 500 MHz to 300 MHz respectively. The output of the diode laser was aligned to the axis of the cavity by first observing the back reflections from the ICOS cavity mirrors, and then tuning the entrance mirror angle while monitoring the transmission signal on a scope. No care was required to stabilize the cavities or the tables upon which they were placed. The cavity was modulated using two methods, which produced identical results. The first method was to modulate the position of one of the cavity mirrors using a piezo driven mount. The second approach was to leave the ICOS cavity mirrors fixed and slightly modulate the angle of injection using a piezo driven final turning mirror. In both cases we employed a commercial piezo driven mirror mount (Thor Labs, KC1-PZ) and applied a low amplitude modulation (1 - 3 volt input to controller, model MDT690) at a sinusoidal frequency of ~1kHz. This modulation resulted in an ICOS cavity coverage of 5 - 10 longitudinal modes. Using this modulation alone it was possible to scan the diode laser and obtain absorption spectra which displayed the expected ICOS intensities, however a noticeable (10%) amplitude modulation persisted. This amplitude modulation was identified as resulting from the ICOS cavity from the frequency spacing, which matched the predicted etalon spacing of the cavity. We believe that this fringing results from the periodic coincidence of the laser frequency with an ICOS cavity mode at the turning points of the piezo modulation. Frequency coincidence at the turning point, where the mirror momentarily stops permits greater transmission into the cavity. This effect is periodic as the laser frequency is scanned.

This weaker modulation was effectively removed through a small modulation in the diode laser frequency over a range that was significantly smaller than the absorption line widths under study. The laser frequency modulation was achieved by applying a 0.1 volt (peak to peak), 150 Hz signal to the Frequency modulation input of the laser, resulting in a 1 GHz frequency modulation. The use of this double modulation was important in realizing a transmission amplitude profile, which was flat with laser frequency. The transmitted signal exiting the rear of the ICOS cavity was directed to an InGaAs detector whose output was digitized directly (no RC filtering) and read into a Labview analysis program which stored the average signal as a function of laser frequency.

Results and Discussion

Spectra of rovibrational overtone-combination bands of water vapor and carbon dioxide were measured, and were used to characterize the performance of the cw-ICOS technique. These measurements were not normalized to variations in the diode laser output. The diode laser exhibited an output power modulation of 1-5% when scanned over spectral regions of greater than several wavenumbers. While this effect could be easily normalized out for wide spectral scans, no attempt was made to do this in the present study since most absorption strength measurements were made on selected lines in regions where the laser output was stable to at least 1 %. In the spectral region between 1.30 and 1.35 micron wavelength, absorptions resulting from H_2O and CO_2 were recorded and compared to known absorption strengths calculated using the HITRAN database [7]. Absorption measurements for single lines were obtained in spectral regions free of laser mode hops, although in some of the wide spectral range data taken of CO_2 some mode hopping is evident. The New Focus diode laser is designed to minimize mode hopping, however there was a noted frequency error between observed and predicted absorption line position near the points of mode hops. This may have been the result of the mechanical grating resetting in the laser head at these points, and could possibly have been reduced by reduction in the frequency scan rate of the system. The absorption data were normalized to the base line cavity transmission level and plotted as fractional absorption as a function of laser frequency.

Absorption due to nascent water vapor in the laboratory air were used in some tests, however the rapidly increasing strength of the absorption for probe wavelengths above 1.35 microns effectively turned the ICOS cell black. Weaker water absorption features near 1.346 microns were used to demonstrate the sensitivity and ease of use of the cw-ICOS technique. A typical 30-cm cavity cw-ICOS absorption spectrum due to atmospheric water is shown in Figure 2, where several absorption features near 7526 cm^{-1} are seen. This spectrum was recorded using the pico-motor scan mode of the New Focus laser. In Figure 2 the absorption spectra predicted using the HITRAN database are shown for a 290 meter pathlength of atmospheric broadened water vapor at a relative pressure of 8.75 Torr (50% humidity at 22 degrees Centigrade). The predicted spectra compares well with the observed spectra in both line width and absorption strength. Examination of the cw-ICOS spectra indicates a fractional absorption of 70% for the line at 7528 cm^{-1},

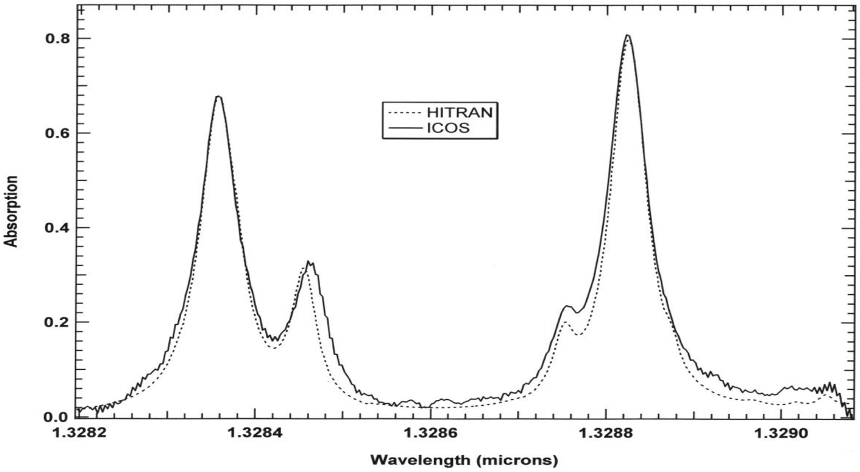

Figure 2. 5 GHz wide scan of three water absorption lines near 7529 cm-1 taken using the cw-ICOS approach. The Y-axis represents fractional transmission through the 30 cm long cavity. The dashed line represents the HITRAN predicted transmission spectrum for a 290 meter long path of air at 50% humidity at 20 degrees Centigrade, at a total pressure of 1 atm.

and a full width at half-max line width of 0.23 cm^{-1}, which agrees well with the HITRAN generated spectra showing a width of 0.2 cm^{-1}. It is important to note that the ultimate resolution that can be realized using the cw-ICOS technique (as described in this paper) is limited by the depth of modulation of the diode laser frequency. In this case, we have utilized frequency modulation of ~1 GHz which should not add significantly to the line width. However it is clear that in order to achieve spectral resolution much greater than this it will be necessary to use much lower frequency modulation (and thus longer ICOS cavities to reduce the mode spacing). The spectral resolution of several GHz offered by this experimental approach represents a good match to the collision broadened line widths seen at atmospheric pressure in the near and mid-infrared spectral regions. This suggests that this approach will be very useful in atmospheric analysis and other monitoring applications. The 290 m path length used in the HITRAN model shown in Figure 2 was selected to give a good line shape comparison. The absolute ICOS signal can be checked using the HITRAN deduced absorption strength and the known cavity length. The predicted line center absorption coefficient for the water absorptions can be deduced from the HITRAN generated spectra. Using the line near 7528 cm-1 a line center absorption strength of $k_a = 4.1\times10^{-5}$ per centimeter is deduced. This results in an ICOS per pass absorption of $1.23\times10^{-3}$for the 30 cm long cavity. Plugging this absorption value into equation 1 where $k\times L = 1.12\times10^{-3}$, $R' = 0.9994 \times \exp[-1.23\times10^{-3}]$, and normalizing to the baseline value where R'=R, yields $I/I_o = 0.30$, which agrees well with the observed absorption peak strength. The weaker absorption peak at 7527.5 cm^{-1} can be similarly analyzed. The HITRAN predicted per-cavity pass absorption factor is 3×10^{-4} resulting in a predicted ICOS cavity transmission 0.60 which is in excellent agreement with the observed absorption signal seen in Figure 2.

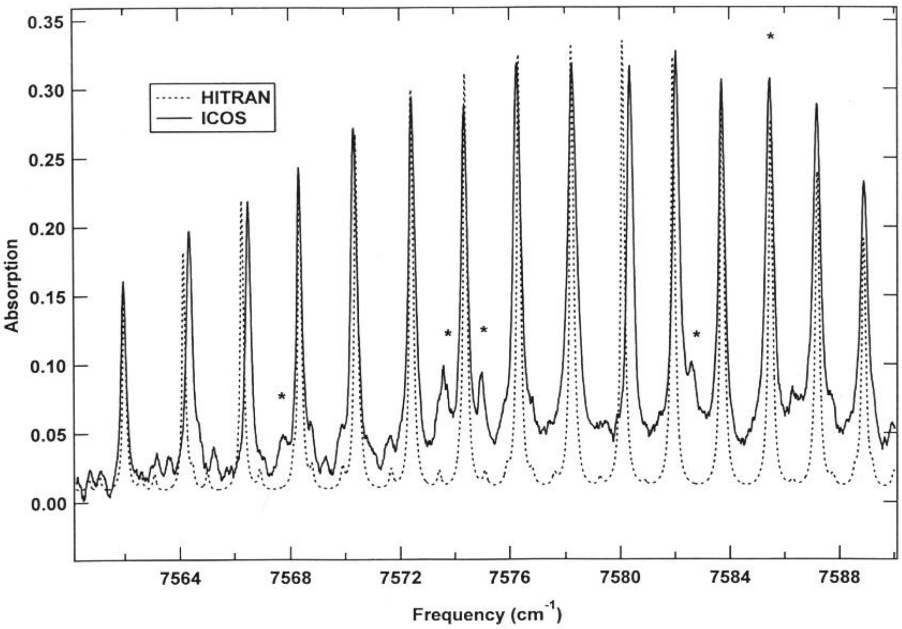

Figure 3) cw-ICOS results recorded using a 1 atmosphere, 50 cm long cell of CO_2 near the overtone band at 7580 cm^{-1}. A HITRAN simulated spectrum corresponding to a 400 meter path, 1 atm sample is overlayed for comparison.

Additional measurements were made using CO_2 as a target gas using a pressure controlled ICOS gas cell. Several overtone/combination bands of CO_2 were measured and compared to the absorption spectra predicted using the HITRAN database. The overtone bands of CO_2 offer a convenient test of the variation of the cw-ICOS signal with intra-cavity absorption strength because the rotational progressions provide a smoothly changing absorption profile to compare with both the impulsive ICOS and coherent Fabry Perot cavity absorption models.

A section of the CO_2 absorption overtone band near 7580 cm^{-1} taken with a 50 cm long cell containing a fraction of an atmosphere of CO_2 is shown in Figure 3, which also shows a overlaid trace of the HITRAN predicted absorption signal for a 400 meter path of 1 atmosphere CO_2. This spectrum was recorded in the fast scanning mode of the New Focus diode laser, which accounts for some of the noise in the amplitude spectra. The excellent match between the HITRAN prediction and the observed cw-ICOS signal clearly demonstrates that the cw-ICOS technique provides an accurate absorption profile of this overtone band, which required only 20 seconds to acquire.The results presented above demonstrate that the cw-ICOS spectra are well described using the “impulsive” ICOS model, as well as the
traditional Fabry Perot absorption models [3,4].

Recently, Engeln, et al. [8] have also demonstrated a somewhat more complex approach to making integrated cavity output spectroscopic measurements, in which a cw laser

spikes fed into a transient recorder which then signal averages the net spectrum as the frequency hopping modes fill the entire scanned frequency range. While these authors did not discuss their Cavity Enhanced Absorption (CEA) work in relation to the well known Fabry Perot absorption models, the underlying optical process is the same, and the absorption "enhancement" resulting from the long cavity residence time is equivalent to that described here and in the earlier ICOS paper [1].

The advantages of the cw-ICOS approach described here are in the system simplicity and ease of application, and the obvious potential for use in a lock-in detection mode of operation. This offers the potential for an increase of several orders of magnitude in sensitivity. We are currently working on this aspect of the technique for applications in quantitative trace analysis .

Conclusions

In this letter, the Integrated Cavity Output absorption method, recently introduced, has been extended to include continuous injection operation and the resulting absorption intensities demonstrated to follow the ICOS model predictions. These results are equivalent to the classic Fabry Perot intra-cavity model predictions (in the short buildup limit). The cw-ICOS approach provides a far easier approach to coupling energy into a high finesse optical cavity, and provides a more generally useful method of making such ultra-sensitive measurements. The use of double modulation, where the ICOS cavity is modulated as well as the frequency of the injection laser, minimizes the transmission amplitude modulation with laser frequency, making this approach possible. Perhaps even more exciting is that the approach can clearly be used in conjunction with post-cavity spectral resolution of broad band width is frequency scanned over a small frequency range (~1 cm^{-1}) and the transmitted cavity mode light sources to provide optical multi-channel array detection of extremely weak absorption [9]. This approach would be of great importance in the concurrent monitoring of several absorbing species.

Acknowledgments

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