Frequency modulation multiplexing for simultaneous detection of multiple gases by use of wavelength modulation spectroscopy with diode lasers

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Modulation frequency multiplexing provides a straightforward method, analogous to television or radio broadcasting, for performing simultaneous detection of multiple gases by use of wavelength modulation spectroscopy with diode lasers. When fiber-optic coupled lasers are used, our approach guarantees that all beams transit the same optical path and impinge on the same detector. Each laser is modulated at a different frequency and the detector output is processed by a set of lock-in amplifiers, one for each laser, to measure the absorbance encountered by each laser. © 1998 Optical Society of America

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Simultaneous or nearly simultaneous detection of multiple gases by use of diode laser spectroscopy usually requires a separate laser for each gas; only in rare, fortuitous cases does one laser exhibit a wavelength tuning range that spans useful absorption lines of more than one gas. The challenge for multiple gas sensing is to combine the light output from several lasers, direct the beams along the same optical path, and perform a separate absorbance measurement for each gas (laser) in a timely manner. We now report simultaneous detection of two gases using wavelength modulation spectroscopy[1–11] (WMS) with two near-infrared diode lasers. Simultaneous detection is possible if one modulates each laser at a different frequency (40 and 50 kHz for the experiments described here) and uses a pair of lock-in amplifiers to process the corresponding WMS signals.

Our approach, called modulation frequency multiplexing, is an improvement over previously described methods for multiple gas detection in which only one laser is activated at a time[12] or the beams are dispersed by a grating onto separate detectors.[13] These previous approaches have obvious limitations. Operating the lasers in sequence[12] obviates simultaneous detection. Although Arroyo *et al*. obtained high repetition rates (1.5 kHz) with their sequential system, their demonstration experiments characterized shock-tube measurements of large, ~30%, absorbances. Single sweep measurements gave adequate signal-to-noise ratios. In contrast, our applications emphasize high sensitivity, long term, continuous detection of weak absorbances.[3] In these applications, true continuous laser operation is preferred in order to implement line locking for wavelength stabilization.[3]

To disperse multiple laser beams with a grating one must add optical paths that differ from gas to gas.[13] Added paths can be problematic when detecting gases that are ubiquitous in air (e.g., water vapor, carbon dioxide, oxygen), particularly when probing small volumes as in the 17.6-cm heated cell that was used for some experiments in Ref. 13. Gratings, because they are diffractive elements, add unwanted optical interference fringes (etalons) that can interfere with detection of weak absorbances. In contrast, modulation frequency multiplexing requires only a single detector and avoids extraneous optical paths and the need for gas purging.

WMS is a well-known method for high sensitivity, optical absorption measurements by use of diode lasers.[1–11] A sinusoidal ac component added to the laser drive current induces a wavelength modulation that is comparable to the width of the absorption feature probed. When the modulated beam is directed through a sample gas and onto a photodiode, absorption by the gas converts some of the wavelength modulation into amplitude modulation, and the induced amplitude modulation appears as ac

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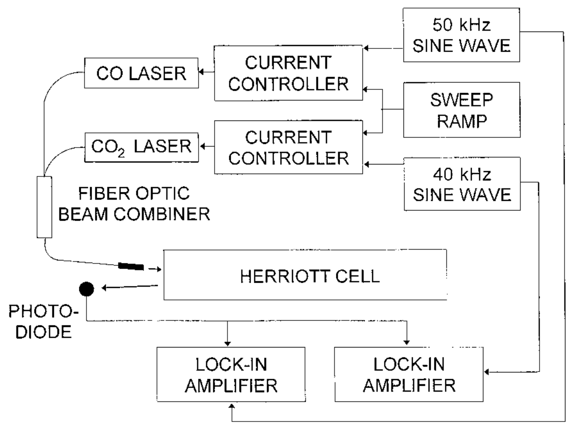

Fig. 1. Schematic diagram of the apparatus used to demonstrate modulation frequency multiplexing for simultaneous detection of carbon monoxide and carbon dioxide.

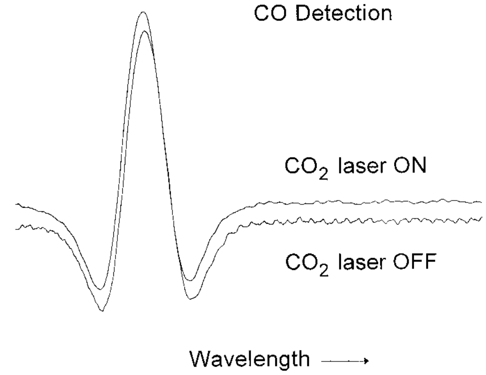

Fig. 2. Modulation frequency multiplexing detection of CO in a 50:50 mixture of CO and CO_2.

components in the photodiode output current. These ac components occur at the modulation frequency and its integer harmonics. A demodulator, such as a lock-in amplifier, can be used to record the magnitude of one of the ac components that is proportional to the optical absorbance by the gas.

Proof-of-principle experiments for modulation frequency multiplexing used two distributed feedback InGaAsP diode lasers: the CO_2 laser manufactured by Northern Telecom has a nominal operating wavelength of 1.548 μm and can be temperature tuned to one of several weak lines in an overtone band of carbon dioxide,[14] the CO laser from Alcatel Optronics operates near 1.558 μm and can be temperature tuned to several high rotational energy lines in the 3 $\leftarrow$ 0 overtone band of carbon monoxide.[14] Each laser is packaged in a hermetically sealed module that contains a miniature thermoelectric cooler, thermistor, monitor photodiode, and optical isolator. Laser light exits the modules through single-mode optical fibers. Temperature regulation and current for each laser are supplied by separate diode laser controllers.

Figure 1 is a schematic diagram of the apparatus used to demonstrate modulation frequency multiplexing for detection of CO and CO_2. The CO laser is modulated at 50 kHz whereas the CO_2 laser is modulated at 40 kHz. One can combine the output from the lasers by using a 2 $\times$ 1 fiber-optic beam combiner and inject it into a single-mode optical fiber. The resulting beam exits the fiber through a collimating graded-index lens and is directed into a Herriott-style[15] multiple pass cell that contains 50 Torr each of CO and CO_2. After traversing 26 passes within the cell (13.9 m), the beam exits and focuses onto an InGaAs photodiode. Output from the photodiode is applied to a pair of lock-in amplifiers set to operate at 2*f*; one is referenced to the 50-kHz modulation source and the other to the 40-kHz source.

We acquired wavelength modulation spectra by sweeping both laser current supplies and digitizing the output from the two lock-in amplifiers. The dc from each supply was adjusted such that a peak that is due to CO appears at the left-hand side of each scan of the CO laser whereas a peak that is due to CO_2 appears at the right-hand side of each scan of the CO_2 laser. This approach—scanning full wavelength spectra and offsetting the positions of the two peaks—is useful for determining signal-to-noise ratios and for examining potential cross talk between the two absorbance measurements.

Key results are displayed in Figs. 2 and 3. Figure 2 compares wavelength modulation spectra when only the CO laser was on (lower trace) and when both lasers were on (upper trace). Spectra were acquired by sweeping the laser current at 39 Hz and coadding 1000 sweeps. The WMS signal corresponds to a peak absorbance of 0.027 (within $\pm$20% of the absorbance predicted by the HITRAN database[14]), and the rms noise (39-Hz detection bandwidth) in the baseline regions corresponds to an absorbance of 0.8×10^{-4} for the lower trace and 1.1×10^{-4} for the upper trace. In contrast, the data in Fig. 3 that show CO_2 spectra indicate that the minimum detectable absorbance actually improves from 1.0×10^{-4} to 0.5×10^{-4} when the CO laser is activated. The CO_2 peak intensity is also within $\pm$20% of the HITRAN prediction.[14]

Examination of the baseline regions in both sets of spectra, Figs. 2 and 3, shows that regular interference fringes dominate random noise. This feature is most clear in the lower trace of Fig. 2. The presence

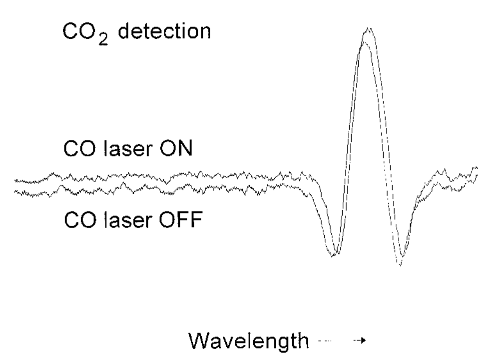

Fig. 3. Modulation frequency multiplexing detection of CO_2 in a 50:50 mixture of CO and CO_2.

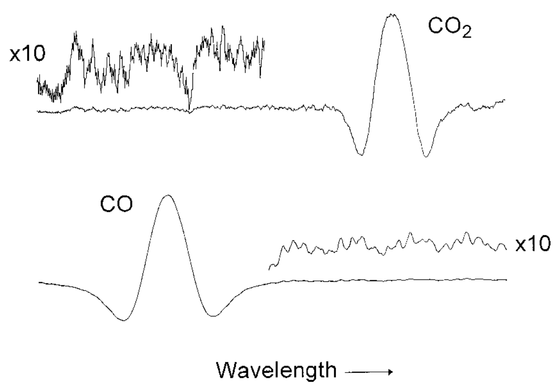

Fig. 4. CO and CO_2 spectra acquired with modulation frequency multiplexing.

of etalons is consistent with our previous experience with nearly all diode-laser-based gas sensing applications and explains why field instruments typically demonstrate minimum detectable absorbances of $\sim 1 \times 10^{-5}$ even though theoretical noise limits are in the low 10^{-8} range. More importantly, the results presented demonstrate no performance penalty associated with frequency-modulated multiplexing. The additional shot noise that is due to having more than one laser impinge onto the detector is overwhelmed by the etalons. Variations in signal-to-noise ratios reported here are due, we infer, exclusively to short-term changes in the interference fringes. Nevertheless, simple numerical filtering of the upper spectrum in Fig. 2 yields baseline noise corresponding to a minimum detectable absorbance below 1×10^{-5} for a 1-Hz effective bandwidth.

Spectra in Fig. 4 do not show measurable cross talk between the two demodulations. The CO and CO_2 spectra were acquired simultaneously using the same conditions as for the spectra in Figs. 2 and 3. There is no excess noise or residual signal that is due to the CO line in the CO_2 trace and vice versa.

Quantitative application of wavelength modulation spectroscopy requires a measurement of the laser power reaching the detector. This measurement can be difficult when more than one laser beam is directed to a single detector. Fortunately, modulation frequency multiplexing allows a simple solution to this problem when combined with near-infrared diode lasers: the amplitude modulation induced by the ac applied to the laser provides a useful measure of the power of each laser beam with the amplitude modulation frequency providing the means for distinguishing the separate contributions.

The multiplexing scheme demonstrated with two lasers can be readily extended to operate many lasers simultaneously. Our approach is analogous to conventional television and radio broadcasting, in which tuned demodulators are used to select among a plethora of signals.

We also note that modulation frequency multiplexing gave better results (higher detection sensitivity) than did a time division multiplexing scheme implemented with an electromechanical fiber-optic switch to select one laser beam at a time. Optical components within the switch increased the size of the unwanted etalons by a factor of 2.5.

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