A Phase-Based Metrology System for Measuring Trace-Gas Concentration

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Project Summary

I. Introduction

Recent advances in tunable laser diodes and their availability at multiple wavelengths in the near-IR has opened a new era of spectroscopic gas concentration measurement techniques. By adjusting temperature and injection current, the laser diode can be tuned to the wavelength of a molecular resonance of interest, and the absorption can be measured and used to obtain concentration. The injection current can also be modulated, allowing for synchronous detection schemes such as wavelength and frequency modulated spectroscopy (WMS and FMS) which effectively bypass the large 1/f noise component of the laser diode. In this paper, a new WMS technique is presented which measures phase due to the anomalous dispersion around a resonance for trace gas detection.

II. Theory

This technique uses a polarization sensitive Mach-Zehnder interferometer to measure a phase signal which is proportional to the slope of the anomalous dispersion profile. One arm of the interferometer contains a sample cell filled with the gas under study and the other arm a vacuum reference cell as shown in Figure 1.

Figure 1.

The laser is dithered about the line center with an amplitude equal to the full-width at half-max (FWHM) of the transition. Light which traverses the sample gas cell develops a phase shift relative to the vacuum reference cell due to the anomalous dispersion in the gas; the resulting optical signals at the detectors are given by

\[ P_{x,y} = \frac{P}{2} (1 \pm k_{yj} \sin(2\pi\sigma L(\Delta n))) \]  

(1)
where $P_0$ is the laser diode power, $k_{1f}$ is a constant gain term associated with harmonic detection, $\sigma$ is the wavenumber, $L$ is the path length, and $\Delta n$ is the refractive index difference due to the anomalous dispersion. The phase is extracted by normalizing the difference signal by the sum signal,

$$V' = \frac{V_{+\Delta} - V_{-\Delta}}{V_{+\Delta} + V_{-\Delta}} = k_{1f} \sin(2\pi\sigma L(\Delta n)).$$

Equation 2 also shows that this technique eliminates laser relative intensity noise (RIN). The refractive index difference, $\Delta n$, for an input frequency modulation equal to the FWHM is

$$\Delta n = \frac{\alpha}{4\pi\sigma_0}.$$  

For ppm and ppb concentrations, the magnitude of the phase becomes

$$V\phi = k_{1f} \frac{\alpha L}{2}.$$ 

Equation 4 demonstrates the direct dependence of phase on absorption and, thus, concentration.

Figure 2 depicts the relationship between slope of the anomalous dispersion and phase modulation amplitude. The input frequency modulation, $\Delta\omega$, generates an output phase modulation, $V\phi$, which is proportional to the slope of the anomalous dispersion. It is also clear from Figure 2 that this phase-based method produces a significantly larger signal than absorption techniques for small modulation amplitudes. This difference is because the slope of the absorption profile goes to zero near line center while the slope of the anomalous dispersion profile is at its maximum at line center.

III. Experimental Results

Measurements have been made using this technique for water vapor at 1392.5 nm. Water vapor cells were made containing two different known concentrations. A low concentration cell was filled with dry nitrogen of a known water quantity and pumped down to the Doppler region. A high concentration cell was filled with 100% RH air and pumped down to the same pressure. A computer model was used to overlay the data and extract a concentration measurement. Figure 3 shows the profile obtained at the first harmonic of the input modulation frequency for the high concentration cell. The profile generated is actually the first derivative of the anomalous dispersion curve as dictated by the Kramers-Kronig relations; the profile seen at the second harmonic is the second derivative, which also resembles the original anomalous dispersion curve. The measured concentration was $3.25 \times 10^{15}$ molecules/cm$^3$ with a theoretical concentration of $9.8 \times 10^{14}$ molecules/cm$^3$. The discrepancy can be attributed to the sealing process of the cells extracting excess water from the cell walls. The low concentration profile is shown in Figure 4. The measured concentration was $6.5 \times 10^{14}$ molecules/cm$^3$ with a theoretical concentration of $2.4 \times 10^{13}$ molecules/cm$^3$. Again, uncertainty in the control measurement can be attributed to the sealing process. The asymmetry in the low concentration profile is the result of a slight path difference in the two arms of
the interferometer which exhibits itself as a bias away from the resonance. This bias was small, however, in comparison to the measurement signal.

Further work demonstrating this measurement technique is in progress utilizing variable pressure cells with a hygrometer, a pressure transducer, and a vacuum pump. The goal is to eliminate the uncertainty due to the sealing process through real time monitoring of the cell’s humidity and pressure. This work will allow for multiple measurement at various concentrations in the Doppler-, Voigt-, and Pressure-broadened regimes.

Figure 3. Figure 4.

IV. Summary

A new and innovative method for trace gas detection has been presented using the slope of the anomalous dispersion curve around a resonance to determine concentration. This method has proven superior to straight absorption techniques in that the measurement signal is independent of laser relative intensity noise as well as intensity modulation. This phase-based technique also results in a significantly larger signal that absorption-based methods for small modulation amplitudes.

References