NEAR-INFRARED DIODE LASER ABSORPTION DIAGNOSTICS FOR TEMPERATURE AND SPECIES IN ENGINES

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October 2004

TSD Report 154

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Abstract

Engine researchers desire spatially and temporally-resolved measurements of temperature and gas species concentrations in the harsh combustion environments associated with modern propulsion and internal combustion devices. The potential benefits of real-time diagnostics are improved engine performance and reduced pollutant emissions. Water vapor has been identified as an attractive species for in situ engine measurements, as it is a major combustion product that may be spectroscopically probed by tunable laser sources.

Near-infrared, fiber-coupled, distributed feedback (DFB) lasers in the 1.31 to 1.47 micron spectral region have been selected to probe water vapor transitions (2ν₂ and ν₁ + ν₃ bands). Direct absorption methods and wavelength modulation absorption spectroscopy have been developed and used to measure temperature and species concentrations in several full-scale engine test facilities as well as in laboratory validation experiments in heated optical cells at low and high pressures.

Several measurement campaigns are described in this work, including a SCRAMJET facility, a high-pressure gas turbine sector rig, and a multi-tube high-repetition-rate pulsed detonation engine (PDE). All three tests have demonstrated the feasibility of making diode-laser measurements in engine facilities with minimal intrusion. These measurement campaigns have provided realistic conditions that have facilitated sensor improvements in optical hardware as well as spectroscopic techniques.

In particular, wavelength modulation spectroscopy (WMS) with second harmonic detection (2f) has been explored as a means to improve temperature measurements in both low-pressure gases with isolated spectral features, as well as in gases with congested
spectra, such as pressure-broadened water vapor. Experiments and analysis, with water vapor at atmospheric pressures and below, show that $2f$ ratio thermometry may be performed over large temperature ranges with minimal calibration and simple interpretation, provided that laser modulation depths are optimized. $2f$ methods not only provide benefits in dealing with weakly-absorbing features, but also demonstrate advantages for diagnostics with broadened and blended spectral features. Experiments and simulations with pressure-broadened water vapor, at pressures up to 20 atm, demonstrate the viability of this strategy. In these situations, $2f$ signal strengths depend critically upon laser modulation depths, which have been increased to values above those typically employed for WMS with diode lasers.
Acknowledgements

This work would not have been possible without the encouragement and insight provided by my advisor, Professor Ron Hanson, as well as the direction and attention to detail offered by Dr. Jay Jeffries. Professor Hanson has helped to elevate my work to a higher level, and has inspired me by his desire for perfection and his incredible industry, especially in laboring over numerous drafts of presentations and manuscripts. I would like to thank Professor Christopher Edwards, as well, for offering to serve on my reading committee alongside Professor Hanson and Dr. Jeffries. Thanks to Professors Thomas Kenny and Richard Zare for serving on my examination committee.

Much help has been provided by fellow diode lab students Xin Zhou, Xiang “Sherry” Liu, Kent Lyle, Adam Klingbeil, and Greg Rieker. The professional and social interactions I’ve had with everyone in the Hanson group have made this journey a great one.

I’d like to thank my church family at PBC as well as my campus fellowship, CCFS, for growing me and supporting me with their deep friendship. I am eternally grateful for my parents, Clark and Diana, who have blessed me with their love, prayers, and wisdom. Thanks also to my sisters Mabel and Nancy, as well as my brother-in-law, Dwight, for their steady love and care. Finally, all this is made possible by the grace of God, to whom this work is ultimately dedicated.

This research was supported in part by a National Science Foundation Graduate Research Fellowship, the Air Force Office of Scientific Research, the Office of Naval Research, and Pratt and Whitney.
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Chapter 1. Introduction

1.1 Background and Motivation

Near-infrared (NIR) diode-laser sensing for combustion applications has been a developing field over the past decade. Water vapor is a strong absorber in the NIR region where mature telecommunications diode lasers are readily available at a variety of wavelengths. For this reason, much of the spectroscopic sensor development that has been done in the near-infrared, including the work done here, has been focused on probing rovibrational water transitions. As this technology continues to improve, an increasing number of engineers and scientists are seeking to utilize these sensors in large-scale engine test facilities for obtaining in situ, spatially and temporally-resolved measurements of parameters such as gas temperature, species concentrations, velocity, or engine-process timing. Real-time engine sensing and control is one ultimate goal, for the purpose of improving engine efficiencies and overall performance, as well as reducing pollutant levels.

Diode laser absorption-based sensors have been researched extensively as a method for making sensitive and fast (kilohertz bandwidth) in situ measurements in combustion environments, as well as for other gaseous or plasma applications such as semiconductor processing (Allen, 1998; Baer et al., 1996; Chou et al., 2001; Hovde et al., 2001; Liu et al., 2002; Philippe and Hanson, 1992; Richter et al., 2000; Silver and Kane, 1999; Teichert et al., 2003). Diode lasers have been used to infer species concentrations, pressures, temperatures, and velocities by rapidly scanning laser radiation
across isolated absorption features and recording highly-resolved absorption line shapes. The use of wavelength-multiplexed diode lasers has shown great advantages for making temperature measurements in hot gases by line-ratio thermometry (Baer et al., 1996; Nagali and Hanson, 1997; Philippe and Hanson, 1992; Sanders et al., 2000; Teichert et al., 2003). In ratio thermometry, multiple diode lasers are used to simultaneously scan across isolated absorption features, or a single laser is used to scan across adjacent absorption features (Silver and Kane, 1999) with line strengths that exhibit different temperature dependencies due to differences in the lower state energies of the chosen absorption transitions. By taking ratios of peak absorbance or ratios of spectrally integrated absorbance (line strength), temperature is inferred while eliminating the need to know species concentrations (Arroyo and Hanson, 1993).

In continuous diode-laser measurements, drifts in laser intensities over time occur due to thermal, mechanical and electronic effects. Therefore, scanned wavelength methods are beneficial because the non-absorbing wings of the spectroscopic features may be used to infer a zero-absorption intensity baseline with each laser scan (usually at kilohertz rates or higher). This assumes that the pressures are low enough, and the absorption lines sufficiently well selected, such that the features of interest are spectrally isolated from neighboring features. For direct absorption measurements with diode lasers, the polynomial fit generally employed to determine the zero-absorption baseline is a critical step in extracting accurate line shapes (Furlong, 1998). Low signal levels and/or various noise sources such as baseline-fitting errors, turbulent flow field-induced beam steering, fiber mode distortion, and etalon effects, often hamper the accurate determination of line shapes.

Wavelength modulation, in conjunction with second-harmonic ($2f$) detection, is well known as a means to increase signal-to-noise ratios (SNR) but at the expense of ease of interpretation and simplicity of design (Silver and Kane, 1999; Bomse et al., 1992). However, for temperature measurements, taking $2f$ peak ratios of absorption features, and making a judicious choice of modulation indices, will be shown to simplify signal interpretation and reduce the need for calibration over large temperature ranges. Also, with the advent of modern computer technologies and software, it is possible to perform wavelength modulation spectroscopy using digital waveform generation and lock-in
detection at hundred-kilohertz rates without dedicated signal generation or lock-in hardware (Fernholtz et al., 2002). Commercial digital lock-ins are available, but generally do not offer the multi-channel capabilities and customization that software lock-ins provide on a PC.

For certain combustion environments, $2f$ spectroscopy is a simple method of yielding temperature measurements with higher accuracy and bandwidth than direct absorption measurements alone (Silver and Kane, 1999). For example, the baseline fitting that is required to extract absorption line shapes in scanned direct-absorption measurements is a large source of uncertainty and error that can be effectively eliminated with $2f$ strategies. Baseline fitting is especially problematic in the case of weak absorption features, for which it is difficult to determine where a zero-absorption baseline begins and ends. At elevated pressures, absorption features often combine and broaden to such an extent that the determination of a laser intensity zero-absorption baseline is not possible, especially when transmission fluctuations occur due to beam steering from thermal gradients or mechanical vibrations. In addition, small changes in the fitted baseline can significantly alter the apparent line strength and shape of a feature. Second-harmonic line shapes improve SNR as well as reduce the sensitivity to baseline fitting errors, since they are sensitive to line shape curvature, making them advantageous in dealing with noisy signals and weak absorption features.

For gases with isolated spectral features (simple molecules at low pressures), this thesis presents a straightforward method of performing and interpreting $2f$ ratios for rapid temperature measurements by using fast-scanning digital electronics and by optimizing modulation indices. In addition to a theoretical discussion of $2f$ peak ratio optimization for temperature measurements, experimental validations of the theory are shown. The optimal selection of modulation indices allows one to relate $2f$ peak ratios directly to line strength ratios over large temperature ranges, thereby minimizing the need for calibration.

Figure 1.1: shows spectral simulations of a segment of the NIR water vapor spectrum ($7178 – 7187$ cm$^{-1}$) at 1000 K at various pressures ranging from 1 to 25 atm. Note that neighboring spectral features interfere at elevated pressures and that the zero-absorption baseline disappears. Except under controlled laboratory environments where
laser transmission fluctuations are suppressed, direct absorption measurements are complicated at high pressures, because of the difficulty in determining a zero-absorption laser intensity baseline. Direct absorption methods generally require knowledge of zero-absorption baseline intensity levels in order to calculate absolute absorption levels. Strategies for measuring or inferring this baseline are well-documented in the literature (Baer et al., 1996; Furlong, 1998). 2\textit{f} methods, however, are based upon line shape curvature, and offer a means to make quantitative measurements with broadened spectra without inferring a zero-intensity baseline. Wavelength modulation provides the additional benefit of improved SNR.

![Figure 1.1: Spectral simulation of 1% water vapor in air; 1 cm path length; $T = 1000$ K.](image)

Unlike the scanned 2\textit{f} method employed for isolated spectral features (Liu, et al., 2004), a fixed-frequency 2\textit{f} strategy is often more appropriate for measurements of blended spectra. With currently-available DFB diode lasers, it is not possible to scan across large blended spectra, such as depicted in Figure 1.1, at rapid rates of tens or hundreds of kHz. Instead, wavelength modulation is used to probe 2\textit{f} peak heights at line-center. Although the 2\textit{f} peak ratios are no longer simply interpreted as being
proportional to line strength ratios, as is the case for isolated spectral features with optimized modulation depth, the $2f$ peak ratios may still be predicted and interpreted by spectral $2f$ simulations. For water vapor, these $2f$ simulations may be based upon the HITRAN2000/HITEMP database (Rothman, 2003) updated with the more accurate line strengths reported in a near-infrared survey of water vapor by Toth (1994). A new version of the HITRAN database incorporates many of the improved line strength values measured by Toth, and is scheduled for release soon. Our lab group has received a pre-release of this new version, HITRAN2004, which we have used in many of our recent simulations. However, it is still important to check these parameters carefully in validation experiments.

![Simulated 2f peak height vs. modulation depth](image)

Figure 1.2: Simulated $2f$ peak height vs. modulation depth ($a$). $T = 1000$ K; 1% water vapor; 7185.6 cm$^{-1}$ feature.

In terms of choosing an appropriate modulation depth (defined in Section 2.2.1), limitations in DFB diode lasers prevent the generation of modulation depths sufficient to optimize the $2f$ signal strength at very high pressures. Obtaining as large a modulation depth as possible is critical since $2f$ signal strengths improve greatly with increasing modulation depth when probing most broadened spectral features with DFB lasers. As an example, Figure 1.2 shows that for a representative water vapor feature in the near-
infrared, the modulation depth required to maximize the 2f signal increases at higher pressures due to collisional broadening of absorption features. The maximum modulation depth that is practically attainable by DFB lasers is a function of modulation frequency, which in turn is determined by the desired sensor bandwidth. These trade-offs will be discussed in a following section.

Various attempts have been made to extract quantitative temperature and species information in combustion gases with diode lasers using modulation techniques. Near-infrared distributed feedback diode lasers are widely available and utilized, as they are relatively inexpensive, robust, and rapidly tunable through injection-current modulation. Typical DFB lasers are conveniently fiber-coupled, provide tens of milliwatts of laser power, and are able to tune over several wavenumbers (cm⁻¹) at kilohertz rates. For low-pressure (atmospheric or less) measurements of molecules such as water vapor and oxygen, DFB lasers may be easily scanned, at kilohertz rates, across isolated spectral features, so that various gas parameters may be inferred from the absorption strength and line shape. Difficulties arise when spectral features are broad, which is often the case for the spectra of larger molecules such as hydrocarbon fuels, as well as at elevated pressures for all molecules due to collisional broadening. 2f spectroscopy is a derivative method that is sensitive to line shape curvature rather than absolute absorption magnitude. The utility of this derivative method for making quantitative and reproducible measurements with broadened spectral features will be demonstrated. The measurement principles of this strategy are introduced in a following section.

1.2 Overview of Thesis

The present chapter contains the background and motivation for this thesis. Since diode-laser sensor research has been ongoing for many years, it is important to have a sense for how this work fits into the developmental chronology of the field, as well as how this effort helps to solve some of the problems associated with previous methods. In particular, Section 1.1 serves to motivate and explain why a large component of this thesis is devoted to describing the development of 2f techniques for thermometry and
species measurements. In fact, other than chapters 4 and 5, this thesis will focus primarily on issues related to wavelength modulation spectroscopy and its applications for gas-phase thermometry and species measurements at low and high-pressures. While direct absorption methods are important, and are employed throughout the measurement campaigns described in chapter 5, there already exists a vast body of literature that details the theory and techniques behind direct-absorption diode-laser spectroscopy (Furlong, 1998; Mihalcea, 1999; Nagali, 1998; Wehe, 2000). Therefore, the contents of this thesis have been restricted to describe new developments or background material necessary for the comprehension or motivation of new concepts.

Chapter 2 starts with a summary of direct absorption strategies but primarily gives an overview of WMS theory, using it to analytically explore low-pressure $2f$ peak ratio thermometry with optimized modulation indices, as well as to describe the details for extending this method for thermometry and concentration measurements with broad and blended spectra. Chapter 3 discusses the experimental details of both fixed-wavelength and scanned-wavelength implementations of the $2f$ sensor, with special focus on the unique challenges and concerns in achieving large modulation depths for high-pressure diagnostics.

Chapter 4 is devoted to outlining the issues behind optical engineering of these sensor systems, with special attention given to the lessons learned from the numerous measurement campaigns described in chapter 5. An excellent spectroscopic technique is of little value if the optical hardware is poorly designed and introduces overwhelming amounts of noise due to beam-steering, fiber mode-noise, or a variety of other factors. Chapter 4 seeks to provide insight into designing an optimized optical system.

Chapter 6 describes the experimental work done to validate the $2f$ measurement techniques introduced in chapters 2 and 3. Low- and high-pressure results for $2f$ ratio thermometry are presented, as well as an example of a high-pressure concentration measurement using $2f$ peak heights.

Chapter 7 summarizes the major findings of this work and suggests some future directions for this research. Appendix A contains data from various validation experiments performed to optimize the spectroscopic line selection for temperature diagnostics using high-pressure $2f$ ratio thermometry. Appendix B contains additional
schematics and photographs of test facilities and the apparatus used on the measurement campaigns described in chapter 5. An alphabetical list of cited references is provided at the conclusion of this report.
Chapter 2. Spectroscopic Theory

The theory of direct-absorption spectroscopy has been described fully by numerous researchers (Furlong, 1998; Mihalcea, 1999; Nagali, 1998; Wehe, 2000) and will not be presented in great detail in this thesis. However, a brief discussion of various direct-absorption strategies, as utilized in our measurement campaigns, will be carried out in Section 2.1. The theory of wavelength modulation spectroscopy is also well-known and well-documented. However, since it may not be as familiar to the average reader, and because it plays a central role in much of the new research developments covered in this thesis, a presentation of the theory of WMS is warranted in Section 2.2.1. That discussion includes a more specific analytical development of the theory for thermometry and species concentration measurements utilizing $2f$ spectroscopy.

2.1 Direct Absorption Strategies

The fundamental equation for absorption spectroscopy is the Beer-Lambert Law. The Beer-Lambert relation gives the transmitted intensity, $I_t$, of monochromatic radiation after passing through an absorbing gas, given here for a uniform gas medium:

$$\tau(\nu) = \left( \frac{I_t}{I_0} \right)_{\nu} = e^{-\alpha(\nu)L} \quad (2.1)$$
τ(ν) is the transmission coefficient, \( I_0 \) is the incident radiation intensity, \( \alpha(ν) \) is the absorption coefficient and \( L \) is the optical absorbing path length. The absorption coefficient is the product of line strength, \( S \) [cm\(^2\) /atm], absorbing species partial pressure, \( p_i \) [atm], and line shape function \( \phi(ν) \) [cm]: \( \alpha(ν) = S \phi(ν) p_i \). The absorption coefficient is a function of temperature, pressure and gas composition, which enables these parameters to be inferred through carefully-designed spectroscopic measurements.

The line strength of an absorption feature is a function of temperature, the transition frequency, \( ν_0 \), the lower-state energy of the quantum transition, \( E'' \), as well as the partition function of the species, \( Q(T) \) (rovibrational states are relevant for near-infrared water vapor spectra):

\[
S(T) = S(T_0) \left( \frac{T_0}{T} \right)^{Q(T)} \frac{1 - \exp \left( - \frac{h \nu_0}{kT} \right)}{1 - \exp \left( - \frac{h \nu_0}{kT} \right)} \exp \left( - \frac{hc}{K} \frac{E''}{T_0} \left( \frac{1}{T} - \frac{1}{T_0} \right) \right)
\]

(2.2)

Here, \( K \) is Boltzmann’s constant, \( S(T_0) \) is the line strength at a reference temperature \( T_0 \) (generally tabulated at room temperature, 296 K), \( h \) is Planck’s constant and \( c \) is the speed of light. The partition function has been analytically determined by various researchers who have provided correlations used in our spectral simulations (Gamache, et al., 2000; Harris, et al., 1998). The correlations provided by these two groups agree well with each other.

The line shape function, \( \phi(ν) \), is determined by a variety of line-broadening mechanisms. There are two main broadening mechanisms that are significant for the measurement environments encountered in this research: Doppler broadening, \( \phi_D \), and collisional (pressure) broadening, \( \phi_C \). A survey of some other broadening mechanisms, which are significant in special cases, may be found in the literature (Wehe, 2000; Demtröder, 1996).

Doppler broadening is due to thermal motion of atoms and molecules, with a velocity distribution described by the Maxwell-Boltzmann expression. The Doppler shift associated with each velocity component may be averaged over the Maxwell-Boltzmann
distribution, resulting in a Gaussian line shape function (2.3). Note that in this expression, the Doppler width, $\Delta \nu_D$, is the half width at half maximum (HWHM) of the Doppler line shape and NOT the full width at half maximum (FWHM). An expression for the Doppler width will be given in Section 2.2.2, in which the implications of these line shapes on wavelength modulation spectroscopy will be discussed.

$$\phi_D(\nu) = \frac{1}{\Delta \nu_D} \left[ 1 + \frac{\ln 2}{\pi} \exp \left[ -4 \ln 2 \left( \frac{\nu - \nu_0}{\Delta \nu_D} \right)^2 \right] \right]^{1/2}$$

Collisional broadening is due to perturbations in the energy levels of absorbing molecules by the electric fields produced by colliding molecules. In its simplest form, which will suffice for this work, the collisional line shape may be approximated by a Lorentzian function (2.4). Note that in this expression, the collisional width, $\Delta \nu_C$, is the HWHM of the Lorentzian line shape and NOT the FWHM. An expression for the collisional width will be given in Section 2.2.2, in which the implications of these line shapes on wavelength modulation spectroscopy will be discussed.

$$\phi_C(\nu) = \frac{1}{\pi} \frac{\Delta \nu_C}{(\nu - \nu_0)^2 + \Delta \nu_C^2}$$

In many situations, Doppler and collisional broadening are both significant contributors to the overall line shape of a spectroscopic feature. The convolution of the Gaussian and Lorentzian line shapes is described by the Voigt line shape (2.5).

$$\phi_V(\nu) = \frac{1}{\Delta \nu_D} \sqrt{\frac{\ln 2}{\pi}} \cdot V(a_{\text{Voigt}}, w)$$

The Voigt function, $V$, is a tabulated or numerically-calculated function (Scheier, 1992; Whiting, 1968) that is a function of the Voigt-$a$ parameter (2.6) and the non-dimensional relative line position, $w$ (2.7). The Voigt-$a$ parameter expresses the relative significance
of the Lorentzian and Doppler broadening mechanisms. For very small or very large values of $a_{\text{Voigt}}$, the Voigt line shape reduces to the Doppler or collisional line shape, respectively.

$$a_{\text{Voigt}} = \frac{\Delta v_c \sqrt{\ln 2}}{\Delta v_D}$$  \hspace{2cm} (2.6)

$$w = \frac{\sqrt{\ln 2} \cdot (v - v_0)}{\Delta v_D}$$  \hspace{2cm} (2.7)

Readers desiring a more detailed and rigorous development of these spectroscopic concepts and relations are encouraged to explore the abundant literature on this topic (Banwell and McCash, 1983; Herzberg, 1944; Herzberg, 1991; Vincenti and Kruger, 1983; Yariv, 1982).

There are two types of direct absorption sensors, those that are fixed in wavelength, and those that involve wavelength scans. Fixed-wavelength sensors are the simplest to operate. Diode lasers are tuned to lase at the peak of an absorption feature, and monitor changes in the peak absorption magnitude, which is a function of temperature and pressure. One advantage of a fixed-wavelength sensor, other than simplicity of design, is that measurement bandwidths are limited by the detector bandwidth alone and may easily reach MHz rates. A major disadvantage of fixed-wavelength sensors is that it is difficult to infer the baseline laser intensity in the absence of absorption, which is necessary to quantify the absorption magnitude. A calibration is required, either with a purged gas sample with no absorbing species, or with a gas sample at a known temperature and pressure, from which the baseline laser intensity may be inferred. However, in any practical combustion environment, transmission fluctuations occur due to beam steering, window fouling, vibrations, etc. In order to account for these transmission fluctuations, a non-resonant (non-absorbing) laser beam is often used as a transmission probe. Unfortunately, multiplexed lasers usually do not track identically with each other, especially in a fiber-coupled configuration. The end result of this is that fixed-wavelength, direct-absorption methods must be calibrated often. Combustion
processes that are periodic in nature, such as an internal combustion engine or a pulsed detonation engine, are therefore more suitable for fixed-wavelength methods, as the sensor may be calibrated at a predetermined point within each engine cycle.

An additional drawback of fixed-wavelength methods is that pressure-shift must be taken into account when measurements are taken over a large range of pressures. The pressure-shift mechanism is a pressure-dependent effect similar to collisional broadening. The line-center position of a spectroscopic feature is slightly shifted in frequency as a function of pressure. If this fact is not taken into account, errors will result from assuming that a fixed-wavelength sensor remains at peak-center as the pressure varies.

Scanned-wavelength, direct-absorption sensors are useful in measurement applications where isolated and narrow spectral features exist. Linear injection current ramps (sawtooth waveforms) are used to scan DFB diode lasers both in intensity as well as in wavelength. Figure 2.1a shows a typical direct-absorption scan using a diode laser, in which the laser intensity changes in response to an injection current ramp. Usually, a polynomial fit to the non-absorbing wings of the absorption feature is used to extrapolate a zero-absorption baseline, with which an absorbance plot of the absorption feature may be constructed, as shown for example in Figure 2.1b.

The benefit of a wavelength scan is that a full absorption line shape is recorded. The spectrally-integrated absorbance (area under the curve in Figure 2.2b) gives a
measurement of absorption strength that is independent of pressure (Baer, et al., 1996). Calibration is unnecessary since the baseline fit provides absolute absorbance information. The integrated absorbance is directly proportional to absorbing species concentration as well as the line strength of the transition. If temperature is known, spectral databases such as HITRAN may be used to calculate line strengths for water vapor transitions. If temperature is unknown, the ratio of integrated absorbance from two appropriately-selected transitions may be used to infer temperature (Arroyo and Hanson, 1993), independent of pressure or species concentration.

As discussed in Section 1.1, direct-absorption methods may be limited by insufficient SNR as well as by problems in determining non-absorbing baseline intensities. These problems hamper the performance of direct-absorption sensors when dealing with weak absorption features as well as broad and blended spectral features (high pressure water vapor). The following sections detail the theory of 2f wavelength modulation spectroscopy for ratio thermometry and species measurements with isolated spectral features as well as congested spectra. An explanation of how these methods help to solve the baseline problem is given in Sections 3.1.2 and 3.2.2. Once again, it is important to understand the motivation for pursuing WMS strategies, and especially how they improve upon the shortcomings of direct-absorption methods (see Section 1.1).

2.2 Wavelength Modulation Spectroscopy

2.2.1 General WMS Theory

The theory of wavelength modulation spectroscopy (WMS) is well documented in the literature (Arndt, 1965; Kluczynski and Axner, 1999; Schilt et al., 2003; Supplee et al., 1994; Wahlquist, 1961; Wilson, 1963), as well as various practical implementations of WMS for gas sensing using diode lasers (Aizawa, 2001; Bomse et al., 1992; Chou et al., 2001; Ebert et al., 2000; Fernholtz et al., 2002; Goldstein et al., 1992; Hovde et al.,
WMS has generally been used to probe relatively narrow and isolated spectral features, but the theory may be extended to deal with broadened spectra. The discussion here will follow the work of Reid and Labrie (1981) as well as the extension to account for amplitude modulation covered by Philippe and Hanson (1993). A more rigorous derivation, which is consistent with the presentation below, has been performed by Kluczynski and Axner (1999).

Assuming that a sinusoidal modulation is riding on a constant D.C. diode laser injection current (or a slowly varying ramp in relation to the sinusoidal modulation frequency), the instantaneous laser frequency, $\nu(t)$, and output intensity, $I_0(t)$, may respectively be expressed as:

$$\nu(t) = \bar{\nu} + a \cdot \cos(\omega_m t)$$  \hspace{1cm} (2.8)

$$I_0(t) = \bar{I}_0 + i_0 \cdot \cos(\omega_m t + \psi)$$  \hspace{1cm} (2.9)

The modulation frequency is given by $\omega_m$, $\psi$ is the phase shift between the intensity modulation and the wavelength modulation, while $a$ and $i_0$ are the amplitudes of modulation around $\bar{\nu}$ and $\bar{I}_0$, which are the slowly varying values of the average wavelength and laser output intensity. Laser intensity and wavelength are assumed to vary linearly with injection current in this analysis, which is an appropriate approximation for the hardware and modulation parameters employed here.

The Beer-Lambert relation has already been introduced in (2.1). In WMS, the transmission coefficient, $\tau(\nu) = \tau(\nu + a \cdot \cos(\omega_m t))$, is a periodic, even function in $\omega_m t$, and can be expanded in a Fourier cosine series,

$$\tau(\nu + a \cdot \cos(\omega_m t)) = \sum_{k=0}^{\infty} H_k(\nu, a) \cos(k\omega_m t)$$  \hspace{1cm} (2.10)

where the functions $H_k(\nu)$ are given as:

$$H_0(\nu, a) = \frac{1}{2\pi} \int_{-\pi}^{\pi} \tau(\nu + a \cdot \cos \theta) d\theta$$  \hspace{1cm} (2.11)
For $2f$ detection, a lock-in amplifier is used to isolate the second harmonic signal. The remaining signal, which contains the second harmonic component of the Fourier sum in (2.10), is given by the relation:

$$S_2(\nu) = -\frac{i_0}{2}H_3(\nu, a) + i_0H_2(\nu, a) - \frac{i_0}{2}H_1(\nu, a)$$

(2.13)

According to Kluczynski and Axner (1999), other harmonics, such as $H_0$ and $H_4$ also couple into Equation 2.13, but are higher-order terms that couple to weak nonlinearities in the laser intensity modulation. Therefore, they have been neglected here. We assume that the phase shift between the laser intensity modulation and the optical frequency modulation is approximately $\pi$ (increasing intensities correspond to decreasing frequencies for DFB lasers – see Section 3.2.3). In addition, these equations do not include the effects of nonlinearities in the modulation, which create harmonic distortions that are manifested as a background $2f$ signal. These residual amplitude modulation (RAM) effects are discussed in detail by Kluczynski and Axner (1999). For the low modulation depths utilized for $2f$ measurements of isolated and narrow water vapor absorption features (atmospheric pressures and below), these effects are not significant. However, for the large modulation depths used to measure pressure-broadened absorption features, these effects are substantial and are discussed in Section 3.3.2.

For optically thin samples, $\alpha(\nu) \cdot L \ll 1$ (or less than 10%), the transmission coefficient reduces to

$$\tau = \exp(-\alpha \cdot L) \approx 1 - \alpha \cdot L = 1 - S \cdot \phi \cdot p_i \cdot L$$

(2.14)

and the second harmonic Fourier component simplifies as:

$$H_2(\nu, a) = - \frac{S \cdot p_i \cdot L}{\pi} \int_{-\pi}^{\pi} \phi(\nu + a \cdot \cos \theta) \cos 2\theta \cdot d\theta$$

(2.15)

Therefore, the $2f$ peak height is only complicated by the line shape function, $\phi$. In order to infer gas temperature by taking ratios of $2f$ peak heights, it is desired that the ratios should only depend upon the well-known line strengths, $S$, of the selected absorption
features. This is possible if the integral in (2.15) is either constant or varies similarly with respect to temperature for each absorption feature, since \( p_i \) and \( L \) cancel in the ratios. This requires wisdom in choosing the modulation index, \( m \), which is defined as:

\[
m = \frac{a}{\Delta \nu}
\]

(2.16)

where \( \Delta \nu \) is the half width at half maximum (HWHM) of the absorption line. Figure 2.2 illustrates how the line-center \( 2f \) peak height varies as a function of \( m \) for Gaussian (Doppler broadened) and Lorentzian (pressure-broadened) line shapes. The functionality is due entirely to the integral in (2.15). The peak value occurs at \( m \sim 2.2 \) for all line shapes, as shown by Reid and Labrie (1981). The second harmonic signal strength that is simulated in Figure 2.2 is calculated by numerically integrating (2.15) for Gaussian line shapes. For Lorentzian line shapes, an exact solution to the integral in (2.15) is used to calculate signal strengths (Arndt, 1965).

![Normalized 2f peak height vs. modulation index, m](image)

Figure 2.2: Normalized 2f peak height vs. \( m \) for a Lorentzian and Gaussian line shape

While the second Fourier component, \( H_2 \), is the dominant term in (2.13), the effects of amplitude modulation are introduced through the higher-order terms containing \( i_0 \). These terms introduce asymmetries into the 2f line shapes. However, since \( H_1 \) and \( H_3 \)
are odd functions that are zero-valued at peak center in the case of isolated and symmetric absorption features, the line-center $2f$ peak height is unaffected by the presence of amplitude modulation distortions. For this reason, measuring line-center $2f$ peak heights offers a simplifying advantage over measuring other parameters such as peak-to-trough amplitudes or full $2f$ line shapes. Figure 2.3 shows simulations of the various terms that contribute to the measured $2f$ signal, $S_2$, in (2.13). For this simulation, $m \sim 2.2$ and the intensity modulation depth was assumed to be 15% ($i_0 = 0.15 \bar{I}_0$), which was typical for the lasers and experimental conditions utilized in this work. The relative magnitudes of the higher-order terms in (2.13) are depicted in this plot, along with the measured $2f$ line shape, which is the sum of the terms. The simulations clearly illustrate that laser-intensity modulation effects do not alter the line-center $2f$ peak heights.

Simulations are performed using the LabVIEW graphical programming language. Absorbance line shapes, $\phi$, are first simulated with an appropriately fine frequency resolution using the HITRAN database, or a corrected-HITRAN database, at the conditions of interest (temperature, pressure, path length, mole fraction, frequency range). Voigt profiles are computed using the numerical approximation given by Whiting et al. (1968). The absorbance spectral simulations are then used to numerically integrate (2.15) and, for odd harmonic terms, (2.12). Since the integration is performed as a function of $\theta$ rather than frequency, the integrands in (2.12) and (2.15) are interpolated from the spectral simulations for each integration step in $\theta$. A trapezoidal rule is used for the actual integration, which is accomplished by a standard LabVIEW integration function.
Figure 2.3: Harmonic components to the $2f$ signal due to laser intensity modulation effects. $m \sim 2.2; i_0 = 0.15 \bar{I}_0$.

Note that in Figure 2.3, the average (D.C.) laser intensity is assumed constant. The asymmetric wings are only due to the fact that the laser intensity is modulated along with the wavelength (as is the case for DFB diode lasers). The asymmetry is NOT due to the fact that the average laser intensity, $\bar{I}_0$, might be slowly changing as the laser scans across the absorption feature. As stated earlier, our analysis here assumes that $\bar{I}_0$ is slowly varying. For scanned-wavelength $2f$ with a DFB laser, as detailed in Section 3.1, $\bar{I}_0$ changes slightly (a linear ramp is typical) while a $2f$ scan is performed across an absorption feature, such that an additional asymmetry may be introduced to the $2f$ line shape. This added asymmetry is due to changes in $\bar{I}_0$ across the feature and NOT because of the laser-intensity modulation effects shown in Figure 2.3.
### 2.2.2 Optimization for low-pressure thermometry

When probing isolated spectral features, it is possible to perform 2f ratio thermometry in an optimized fashion such that the 2f ratios are proportional to line strength ratios over large temperature ranges. The discussion that follows illustrates this concept.

Temperature measurements using 2f spectroscopy are complicated by the fact that the modulation index, $m$, varies with temperature through $\Delta \nu$ in (2.16). Doppler (Gaussian) line widths vary with the square root of $T$ whereas collisional (Lorentzian) widths vary with $T$ according to an inverse power law:

$$
\Delta \nu_D = 3.581 \times 10^{-7} \nu_0 \sqrt{T/M} \tag{2.17}
$$

$$
\Delta \nu_L = P \sum_j \chi_j \gamma_j^{T_0} \left( \frac{T_0}{T} \right)^{n_j} \tag{2.18}
$$

In (2.17) and (2.18), $\Delta \nu_D$ and $\Delta \nu_L$ are the Doppler and Lorentzian half widths at half maximum (HWHM), respectively, in wavenumber (cm$^{-1}$) units. $\nu_0$ is the line-center position of the feature in cm$^{-1}$, $T$ is the temperature in Kelvin, and $M$ is the atomic mass (a.m.u.). $P$ is the total pressure in atmospheres, $\chi_j$ is the mole fraction of the $j^{th}$ component of the gas mixture, $\gamma_j^{T_0}$ is the pressure broadening coefficient (half width) at reference temperature $T_0$ for the $j^{th}$ perturbing species (cm$^{-1}$/atm), and $n_j$ is the species-dependent temperature coefficient.

For Doppler-broadened line shapes, the line shape function, $\phi$, only varies with temperature and is nearly identical for all absorption features probed by a multiplexed beam (assuming similar values of $\nu_0$). Therefore, choosing identical modulation indices, $m$, for each absorption feature insures that the integral in (2.15) cancels when taking 2f peak ratios. For pressure-broadened lines, the Lorentzian line shape function may have a slightly different temperature and pressure dependence for each absorption line due to differing broadening parameters, $\gamma_j^{T_0}$ and $n_j$. Also, spectral line widths are dependent upon gas composition, since broadening parameters are species-specific. However, by
choosing identical values of $m$ for each absorption feature near the conditions $(P, T, \chi_j)$ of interest, the $2f$ peak ratios of pressure-broadened lines should be relatively insensitive to differences in the integral in (2.15) for modest temperature, pressure and compositional ranges. These effects may be analyzed and simulated by using parameters found in spectral databases such as HITRAN and HITEMP. The most ideal situation, clearly, is to tune the modulation amplitude such that $m$ is near a value of 2.2 so that the integral in (2.15) is maximized and therefore insensitive to temperature, pressure and compositional effects for each individual absorption feature.

One way to understand this optimization strategy is to look at the mathematical expression for a $2f$ peak height ratio (2.19). When the ratio of the integrals in (2.15) remains constant, the $2f$ peak height ratios are directly proportional to line strength ratio (2.19). Since line strengths are relatively well-known, as a function of temperature, the result is a simply-interpreted sensor strategy requiring minimal calibration (just one point).

$$
\frac{H_2(v_{\text{line}1}, a)}{H_2(v_{\text{line}2}, a)} = \frac{S_{\text{line}1}}{S_{\text{line}2}} \int_{-\pi}^{\pi} \phi(v_{\text{line}1} + a \cdot \cos \theta) \cos 2\theta \cdot d\theta \propto \frac{S_{\text{line}1}}{S_{\text{line}2}}
$$

(2.19)

Since analytical solutions to (2.15) exist in the literature for Lorentzian line shapes (Arndt, 1965; Wahlquist, 1961), it is possible to derive an expression for pressure-broadened $2f$ peak-height ratios provided that the assumption of weak absorption holds. From the work of Arndt (1965), the $2f$ peak height for a Lorentzian absorption feature, identified with the subscript $i$, may be written as:

$$
H_2(v_{0,i}) \propto S_i p L \frac{2}{m_i^2} \left( 2 - \frac{2 + m_i^2}{\sqrt{1 + m_i^2}} \right)
$$

(2.20)

Taking the $2f$ peak ratio of two multiplexed lines, identified with subscripts $i = 1$ and 2, yields:

$$
R = \frac{S_1}{S_2} \left( \frac{2\sqrt{1 + m_1^2} - 2 - m_1^2}{2\sqrt{1 + m_2^2} - 2 - m_2^2} \right) \left( \frac{m_2^2 \sqrt{1 + m_2^2}}{m_1^2 \sqrt{1 + m_1^2}} \right)
$$

(2.21)
From (2.21), it is evident that the 2\(f\) peak ratios are equivalent to line strength ratios if the modulation indices are equal. This is true for two pure Gaussian line shapes as well, but is not true for two arbitrary Voigt profiles unless they have identical parameters. This can be seen by inspection of Figure 2.2, from which we note that 2\(f\) peak heights for all Gaussian and Lorentzian line shapes behave identically with \(m\), because the line shape integrals in (2.15) are identical. However, the behavior of 2\(f\) peak heights for Voigt line shapes fall anywhere between the two limiting cases shown in Figure 2.2. In practice, multiplexed absorption lines that exhibit Voigt line shapes may possess different pressure broadening coefficients, which will result in a mix of Voigt profiles. Therefore, either a numerical integration of the Voigt line shapes (if accurately known), or a calibration near the measurement conditions allows for a determination of a scaling factor between 2\(f\) peak ratios and line strength ratios. In addition, it is often difficult to completely account for differences in laser intensities and lock-in settings, as well as slight differences in \(m\), so that a calibration is generally required when performing modulation spectroscopy.

The data in Figure 2.2 indicate that the 2\(f\) peak signal strength varies quite slowly for modulation indices near 2.2. For example, consider the 2\(f\) peak height for a Lorentzian absorption feature with a temperature coefficient, \(n = 0.5\), at 1600 K, modulated with an optimal modulation index of 2.2. First, using (2.20) or Figure 2.2, the range in which the modulation index, \(m\), is within 1% of its optimal value at \(m \sim 2.2\) is calculated to be \(1.90 < m < 2.55\). This range is used, together with (2.16) and (2.18), to calculate the corresponding temperature range in which the 2\(f\) peak intensity of this particular absorption feature varies with temperature precisely as its line strength does to within an error of 1%. For this Lorentzian feature with \(n = 0.5\), the temperature range is \(1190 \text{ K} < T < 2145 \text{ K}\). For a multiplexed temperature sensor probing two absorption features, this temperature range of nearly 1000 K represents a lower bound because taking peak ratios tends to mitigate the effects of changes in \(m\) with temperature, assuming that both \(m\) values are identical and optimal (\(m \sim 2.2\)). This back-of-the-envelope calculation provides additional assurance that 2\(f\) peak ratios may be assumed to vary directly with line strength ratios, for temperature ranges on the order of 1000 K (at combustion temperatures), provided that modulation indices are optimized.
Figure 2.4 is a plot of optimal modulation depth, $a$ (where $m \sim 2.2$), for which the $2f$ peak height is maximized, as a function of total pressure for a water vapor feature at 1392 nm (7185.6 cm$^{-1}$). The simulation was performed using spectroscopic parameters from HITAN2000, specifying a temperature of 1000 K, a path length of 1 cm and a mole fraction of 1%. For low pressures, the line width is dominated by Doppler broadening, which is a constant value at the specified temperature of 1000 K. As pressure increases, collisional broadening of the line is linear with pressure, which is reflected in the roughly linear growth of $a$ in Figure 2.4. Interference from neighboring features becomes more prominent at high pressures, affecting the linearity of $a$ with pressure. At sufficiently high pressures (5 atm and above), it becomes difficult to define a modulation index, $m$, due to the fact that spectral features are blended rather than comprised of isolated lines. Since line widths are ill-defined for these broadened and often asymmetric features, the modulation index is not a useful concept at high pressures.

\begin{figure}[h]
\centering
\includegraphics[width=0.7\textwidth]{figure2.4}
\caption{Optimal modulation depth, $a$ (where $m \sim 2.2$) as a function of total pressure. Water vapor feature at 1392 nm (7185.6 cm$^{-1}$), $T = 1000$ K, $\chi = 1\%$, $L = 1$ cm. Simulation is based on HITRAN2000.}
\end{figure}
2.2.3 Thermometry for blended and broadened spectra

For Gaussian, Lorentzian, or Voigt line shapes, numerical integration of (2.15) may be performed (Wilson, 1963). In the case of highly broadened and blended line shapes, spectral simulation programs may be used to generate tabular absorbance data, which are readily integrated numerically to simulate $2f$ signal intensities.

A single-point calibration at one known measurement condition is sufficient to scale the measured $2f$ ratios with the simulations for the purposes of thermometry. It is necessary to have an independent measurement of total gas pressure, since the temperature sensitivity of the $2f$ peak ratios is also a function of pressure. By simulating the $2f$ peak heights, and thus the ratios, for two or more water features, at a matrix of relevant pressure and temperature conditions, a single calibration allows for temperature measurements even under variable pressure conditions. However, an independent pressure measurement is required. The results of this study indicate that $2f$ simulations are capable of predicting and interpreting experimental results, but that more detailed spectroscopic studies are necessary in order to insure completely accurate large-modulation-depth $2f$ simulations. These $2f$ simulations are sensitive to parameters, such as broadening widths and temperature coefficients, which have not been experimentally verified with great care in the literature. In Appendix A, results from preliminary direct-absorption and $2f$ validations experiments are shown in an effort to identify good candidate water transitions for high-pressure $2f$ thermometry. Since errors are common in spectroscopic databases such as HITRAN, it is important to perform these rough assessments before settling on a new sensor line choice.

2.2.4 Extracting Species Concentration

When $2f$ ratio thermometry is successfully implemented, a determination of species concentration can also be made. If the total pressure is measured and temperature is inferred using ratio thermometry, the absolute magnitude of the individual $2f$ signals
are proportional to species concentration in the low-concentration, low-absorbance limit (less than 10% absorbance). A one-point calibration at a measurement condition of known $T$, $P$, and mole fraction is, of course, necessary to scale the measurements with $2f$ simulations.

Figure 2.5 shows a $2f$ peak height simulation (for 1% water vapor in air) versus temperature and pressure for a water vapor feature at 7185.6 cm$^{-1}$ (1392 nm) at a modulation depth of 0.5 cm$^{-1}$ and a path length of 1 cm. If gas temperatures and pressures are fluctuating in time across a measurement path, the plot in Figure 2.5 may be used to predict how the $2f$ signal should change if the mole fraction is constant. Since Figure 2.5 simulates how the $2f$ signal changes with temperature and pressure for a constant water mole fraction, it can also be used to infer water mole fraction variations if pressure is known from an external pressure transducer reading, and temperature is known from the method described in the previous section. The premise for this mole fraction inference is that the $2f$ signal is directly proportional to species concentration, which is true for optically thin conditions (less than 10% absorption) and for dilute samples that are dominated by air-broadening, so that changes in mixture composition have a negligible effect on line-shape broadening.
Figure 2.5: 2f peak height vs. temperature and pressure (for 1% water vapor in air). 1392 nm (7185.6 cm$^{-1}$) water vapor feature; $a = 0.5$ cm$^{-1}$, 1 cm path length.
Chapter 3. Experimental Considerations

3.1 Scanned-Wavelength Sensors

3.1.1 Hardware

For scanned-wavelength temperature and concentration sensors, fiber-coupled, distributed-feedback (DFB) diode lasers (10-30 mW), from NEL America Inc., are multiplexed using standard single-mode fiber combiners (1x2, 1x3, or 1x4) designed for the near-infrared. The laser packages (14-pin butterfly) are placed in ILX Lightwave mounts (LDM-4984) and are driven with an ILX Lightwave modular diode-laser controller (LDC-3908) equipped with 500mA/9W current source and temperature control units (LDC-3916372). The lasers are temperature tuned to lase at wavelengths near the selected absorption features, and are set with a constant bias injection current of about 70 mA. Since the NEL lasers are specified to operate with a maximum current level of 150 mA, the 70 mA current level biases the lasers near the midpoint. Higher current settings are employed if needed, as described for the high-pressure work in Section 3.2.1, but are not necessary for low-pressure diagnostics. An external modulation, either consisting of a 1 kHz saw tooth ramp, for direct absorption scans, or a 1 kHz ramp summed with a faster 170 kHz sinusoidal modulation, for scanned-2f spectroscopy, is fed into each of the current-source units. The two waveforms, with independent amplitude control, are generated using LabVIEW codes running a National Instruments data acquisition (NI-
DAQ) system. The NI-DAQ system consists of a personal computer outfitted with a PCI-6115 DAQ board (12-bit A/D conversion) and a BNC-2110 analog I/O block.

The fiber-coupled (9 micron single-mode fiber) multiplexed beam is transmitted across a test region using a Thorlabs aspheric collimator (F230FC-C). On the collection side, a larger collimator from Oz Optics (HPUCO-25-1300-M-10BQ) is used to focus the free space beam into a 400 micron diameter multimode fiber with a numerical aperture (NA) of 0.39 (Thorlabs M20L05). The 400-micron fiber routes the collected signal into a grating-based demultiplexing setup, where a large aspheric lens (Optosigma 023-2392) is used to collimate the output from the 400-micron fiber and send it onto a 30 mm square diffraction grating (1st order) with 1200 grooves/mm (Edmund Optics NT43-852). The demultiplexed beams are then sufficiently dispersed in space to be focused with concave mirrors onto individual 10 MHz silicon photodetectors (Thorlabs PDA400). Care is taken to carefully focus and align the separated beams onto the 1 mm square detector surfaces in order to avoid mode noise distortions caused by the 400-micron fiber. Figure 3.1 illustrates the experimental setup and Figure 3.2 is a photograph of a dispersion setup for demultiplexing three laser channels.

Figure 3.1: Experimental setup.
Detector signals from both channels are simultaneously recorded at a 5 MHz sampling rate by the NI-DAQ system using a LabVIEW scope program. \(2\ell\) line shapes are recovered by running a digital lock-in program on LabVIEW with a low-pass filter time constant of 50 kHz.

3.1.2 Problems with Direct Absorption Scans

Figure 3.3a shows a plot of the detector signal for a single absorption scan taken at a scan rate of 1 kHz. The feature is a weakly-absorbing water vapor line in the NIR at room temperature and atmospheric pressure. Note that laser-intensity noise limits the detection sensitivity to about 0.1% absorbance. A polynomial baseline fit to the non-absorbing wings of the absorption feature is performed to extrapolate a zero-absorption baseline intensity level for the purpose of creating the absorbance plot in Figure 3.3b.
This baseline fit introduces some error to the line shape of Figure 3.3b, especially when the feature is weak (as in this example) or if the feature is broadened by collisions at higher pressures. For broad spectral features, it is difficult to know where the non-absorbing wings of a spectral feature begin and end. Incorrect baseline fits that exaggerate or diminish the apparent absorption magnitude of spectral features are common sources of measurement error. As previously shown in Figure 1.1, at sufficiently-high pressures, non-absorbing baselines completely disappear, adding additional complexity to scanned direct-absorption measurements. As discussed in Section 1.1, $2f$ spectroscopy is a derivative method that is sensitive to line-shape curvature (the $2^{nd}$ derivative) rather than absolute absorption magnitude. Therefore, spectral lines may be quantified by their curvature, even at high pressures.

![Figure 3.3a](Image)

Figure 3.3a (left): Direct absorption scan for a small (1.2% absorbance) water feature, a single scan at 1 kHz, $\lambda = 1392$ nm.
Figure 3.3b (right): Absorbance plot of Figure 3.3a, showing direct absorption single-scan detection limit (1 kHz scan rate), $\lambda = 1392$ nm.

A comparison of signal-to-noise ratios for scanned direct absorption and scanned $2f$ are shown in Figures 3.4a and 3.4b, respectively. A factor-of-five improvement in SNR is achieved, due to the fact that phase-sensitive detection is performed at a higher frequency (50 kHz), thereby reducing $1/f$ laser-intensity noise.
3.1.2 WMS: Adjusting for Transmission Fluctuations

In order to account for wavelength-dependent fluctuations in laser intensity, $I_0$, caused by thermal and mechanical transients, fiber vibrations and changes in fiber positioning (which affect each laser differently), the value of the $2f$ peak signals at line center are adjusted by dividing them by the corresponding line center values of $\bar{I}_0$. $\bar{I}_0$ values are calculated by performing polynomial fits to the non-absorbing portions of the laser scans. The line-center value of $\bar{I}_0$ is then simply inferred from the polynomial fit. As mentioned, the $2f$ peak heights, determined using a LabVIEW peak detector routine, are then divided by the fitted $\bar{I}_0$ value in order to account for laser intensity fluctuations. The peak detector routine uses an algorithm that fits a quadratic polynomial to sequential groups of data points. Due to the fact that the lock-in low-pass filter smooths the $2f$ line shape, the peak detector routine does not introduce significant errors to the measurement. Figure 3.5 is an example of a laser scan, with a fit for $\bar{I}_0$ illustrated as well. The corresponding $2f$ line shape for the signal in Figure 3.5 is shown in Figure 3.6. It is important to note that the error associated with performing polynomial fits to infer $\bar{I}_0$ in our procedure is minimal compared to the error introduced by performing polynomial fits.
to determine direct absorption baselines for weakly-absorbing features. This is due to a
difference in relative measurement scales. A 1% error in $I_0$ adds a 1% error to our
adjusted $2f$ peak heights. However, for a direct absorption measurement, a 1% error in
the baseline fit for a 1% absorption feature (for example) can create a 100% measurement
error.

![Figure 3.5: Example of a modulated laser scan. A fit for $I_0$ is depicted, which is used to
adjust for laser transmission fluctuations.](image)

The ability to correct $2f$ peak heights to account for fluctuations in $\bar{I}_0$ is the major
advantage of a scanned-wavelength implementation as opposed to a fixed-wavelength
method. While fixing the lasers at line center would allow for measurement bandwidths
of 50 kHz (the lock-in time constant) as opposed to the scan-rate-limited 1 kHz
bandwidth of our present set-up, there would be no information on $I_0$ since only $I_t$ is
measured in (2.1). The use of free-space optics, in a well-controlled laboratory
environment, offers a means to circumvent this problem by using beam splitters to
provide $I_0$ reference signals prior to the combustion region. In practice, however, it is
difficult for reference signals to account for uncertainties and changes in transmission
characteristics through a combustor, especially if fiber optic collection is employed. For very low absorbance levels (optically-thin), on the order of 1% or less, fixed-wavelength methods may be feasible since $I_0$ and $I_t$ are nearly equal. Therefore, only a small error, on the order of the absorbance (1%), is introduced if $I_t$ is assumed to be identical with $I_0$. There is much additional literature that recognizes the need for transmission corrections when making WMS measurements (Silver and Kane, 1999; Fernholtz et al., 2002).

![Figure 3.6: Example 2f line shape at 1392 nm. Ambient water vapor at 296 K, 10 cm path length.](image)

### 3.2 Fixed-Wavelength Sensors

#### 3.2.1 Hardware

For fixed-wavelength direct absorption measurements, the temperature and injection current settings are adjusted to tune each laser to the peak of a relevant absorption feature. A non-resonant (non-absorbing) beam is multiplexed with several resonant lasers in order to act as a transmission probe (see Section 3.2.2). This non-
resonant beam is particularly useful for removing high-frequency noise sources such as flow-field induced beam steering and vibrations. However, low-frequency drifts in laser intensities can also occur and these must be accounted for. This is the primary detriment to a fixed-wavelength strategy: namely, the inability to tune off of absorption transitions in order to obtain the laser intensity baseline, $I_0$. In our research, fixed-wavelength strategies are only employed in applications that are periodic in nature, in which there exists a calibration point of a known temperature and pressure condition. For example, in a pulsed detonation engine, the temperature of the fuel plus buffer air mixture at the beginning of each cycle is known and may be used as a calibration point for the sensor.

For fixed-wavelength 2f spectroscopy, the lasers are temperature tuned to lase at wavelengths near selected absorption features, and are set with a constant bias injection current of about 100 mA. The lasers are constrained by the current source to operate with a maximum current level of 200 mA, so that the 100 mA injection current biases the lasers near the midpoint. The 200 mA current level limit, which is higher than the 150 mA specification given by the manufacturer, enables the laser to be modulated with large modulation depths. No degradation in laser performance is observed under these conditions, but higher current limits are not utilized in order to avoid permanent laser damage. An external (typically 50 kHz) sinusoidal modulation is fed into the current-source units. The modulation waveform is generated using LabVIEW codes running a National Instruments data acquisition (NI-DAQ) system. The NI-DAQ system consists of a personal computer outfitted with a PCI-6115 DAQ board (12-bit A/D conversion).

An example of a fixed-wavelength 2f measurement is shown in Figure 3.7. The measurement condition for this experiment was 10 atm and 500 K, with a peak absorbance of only 0.8%. The feature used in Figure 3.7 is the same feature, at 7185.6 cm$^{-1}$ (1392 nm), shown in Figure 1.1. Note that at 10 atm, collisional broadening causes enough interference from neighboring features to eliminate the non-absorbing wings of all features in Figure 1.1, making scanned direct-absorption measurements difficult to interpret. The direct-absorption detection limit, ignoring errors from fluctuations in laser intensity baselines, is approximately 0.1% at 1 kHz. This would result in a direct-absorption SNR of 8 for this 0.8% absorption feature (once again, optimistically assuming that the baseline intensity is stable and known). Figure 3.7 shows an order-of-
magnitude improvement in SNR using 2f methods (SNR ~ 75 over 1 ms). The next section describes how a fixed-wavelength 2f strategy may offer benefits for solving the baseline-fluctuation problem inherent in fixed-wavelength methods.

Figure 3.7: Line-center 2f signal for a water vapor feature at 7185.6 cm\(^{-1}\) (1392 nm). \(T = 50\ \text{atm},\ P = 10\ \text{atm},\ a = 0.5\ \text{cm}^{-1}\)

3.2.2 WMS: Adjusting for Transmission Fluctuations

One of the benefits of a scanned 2f method, as adopted for low-pressure thermometry, is that the laser intensity in the absence of absorption, \(I_0\), may be determined by performing a polynomial fit to the non-absorbing wings of an isolated transition (Section 3.1.2). For broad and blended spectral features that do not possess non-absorbing wings, the scanned 2f method is not advantageous and a fixed-wavelength method is preferred. The benefit of the fixed-wavelength approach is higher measurement bandwidth, but it is more difficult to account for laser transmission fluctuations.

In practice, one method for overcoming this difficulty is by working with optically thin absorption transitions. In this case, \(I\) and \(\bar{I}_0\) may be assumed to be equivalent, so that the average laser intensity may be measured and used to correct for any transmission variations, due to beam steering, mechanical misalignments, soot,
window fouling, etc. An alternative to measuring the average laser intensity is to record the $1f$ component of the modulated signal as a transmission probe (Fernholtz et al., 2002). This $1f$ signal is due to the laser-intensity modulation present in DFB lasers, and is proportional to the average laser power. The optically-thin assumption is necessary, just as when using the average transmitted intensity. Note that this simplification for optically-thin samples does not pertain to direct-absorption measurements. For the same reason given in Section 3.1.2, direct-absorption measurements must resolve small changes in absorption in order to be meaningful, NOT assume that they are negligible.

If the optically-thin limit is not feasible for practical reasons or for SNR requirements, a reference beam may be necessary to monitor incident laser power prior to the absorption path length. The reference beam will account for laser drift and any perturbation before the absorbing gas region, but will not compensate for gas sample-induced effects such as beam-steering, window fouling, soot, etc. Another tactic is to use a non-resonant beam multiplexed with the resonant laser beams to act as a transmission probe. Care must be taken to insure that the transmission characteristic of the non-resonant beam matches the resonant beams, and that all transmission fluctuations, excluding gas absorption, affect all lasers equally.

### 3.2.3 WMS: Characterizing Modulation Depths

In order to predict and compare $2f$ measurements and simulations, an accurate knowledge of the modulation depth is required. In order to characterize the modulation depth of the lasers as a function of the amplitude of the sinusoidal driving waveform, a fiber ring etalon with a free spectral range (FSR) of 0.0277 cm$^{-1}$ is used (E-TEK Dynamics, Inc. model SWBCSNA1PH211). Since the tuning range of the lasers are highly dependent on modulation frequency, it is necessary to perform the characterizations at the exact modulation settings used. At a 50 kHz modulation frequency, a fast digital oscilloscope (Tektronix TDS 3032) is required to resolve the narrow etalon peaks on the sinusoidal waveform. Figure 3.8 shows an example
waveform where etalon peaks are clearly visible and may be counted to determine the peak-to-peak tuning range of the laser at this particular modulation setting. Note that the modulation depth is defined as half the peak-to-peak tuning range of the laser. A plot of modulation depth versus the driving waveform amplitude, as shown in Figure 3.9, indicates that the response is linear and limited by the 200 mA maximum current setting imposed by the current source. Maximum currents of greater than 200 mA were not attempted to avoid damaging the lasers. The manufacturer’s specification for maximum current is 150 mA.

![Etalon fringes on a sinusoidal waveform, for determining laser modulation depth.](image)

$\alpha \sim \frac{29}{2}(0.0277 \text{ cm}^{-1}) = 0.40 \text{ cm}^{-1}$

29 fringes

Etalon free spectral range (FSR) = $0.0277 \text{ cm}^{-1}$

Time axis

$\alpha \sim \frac{29}{2}(0.0277 \text{ cm}^{-1}) = 0.40 \text{ cm}^{-1}$

Detector signal (V)

Figure 3.8: Etalon fringes on a sinusoidal waveform, for determining laser modulation depth.
In many practical devices in which diode-laser-based gas sensors would be a valuable tool, optical path lengths are highly limited. 2\(f\) signal strengths depend critically on laser modulation depth, as shown previously in Figure 1.2. This figure demonstrates that for water vapor, there is a dramatic increase in the modulation depth necessary to maximize 2\(f\) signal strengths as pressures increase. The DFB diode lasers used in this work are generally not modulated more than a few tenths of a wavenumber at the high modulation frequencies employed here (50-170 kHz). Clearly, increasing this parameter is of utmost value for achieving high SNR data from small measurement paths if high-pressure environments are to be encountered. The practical limitation in modulation depth is determined by the diode laser characteristics of tuning rate and maximum current limit. However, there are certain negative side-effects that complicate the
implementation of large-modulation-depth 2f spectroscopy in practice, and which must be resolved in order for this technique to succeed. These concerns, such as intensity modulation effects and laser harmonic distortion, are addressed in the following sections. In addition, modulation depth is a function of modulation frequency, which ultimately has a bearing upon sensor bandwidth and noise, as described in Section 3.3.3.

3.3.1 Intensity Modulation Effects

One of the major benefits of measuring 2f peak heights, as opposed to other 2f waveform parameters such as wing amplitudes, is that laser intensity modulation effects are minimized. The following relations define the wavelength modulation (3.1) as well as the laser intensity modulation in a diode laser (3.2):

\[ v(t) = \bar{v} + a \cdot \cos(\omega t) \]  
\[ I_0(t) = \bar{I}_0 + i_0 \cdot \cos(\omega t + \psi) \]

Here it is assumed that a sinusoidal modulation is riding on a constant D.C. diode laser injection current where \( v(t) \) is the instantaneous laser frequency and \( I_0(t) \) is the output intensity. The modulation frequency is given by \( \omega \) (radians/second) where \( \omega = 2\pi f \) and \( f \) is the frequency in Hertz. \( \psi \) is the phase shift between the intensity modulation and the wavelength modulation, while \( a \) and \( i_0 \) are the amplitudes of modulation around \( \bar{v} \) and \( \bar{I}_0 \), which are the average wavelength and injection current.

The effect of intensity modulation on 2f signals, which is considerable for injection-current-tuned DFB lasers, especially at the large modulation depths, \( a \), employed here, is described briefly in Section 2.2.1 as well as in the literature (Liu et al., 2004a; Philippe and Hanson, 1993; Kluczynski and Axner, 1999; Schilt et al., 2003). In Figure 3.10, these effects have been simulated for a typical DFB diode laser used in our studies. The simulated conditions are of water vapor at 500 K and 10 atm, with a modulation amplitude, \( a \), of 0.5 cm\(^{-1}\), and a representative intensity modulation in which \( i_0 \) is 2/3 of the average intensity, \( \bar{I}_0 \). While the wings of the 2f waveforms are greatly
distorted and are asymmetric due to amplitude modulation effects, the $2f$ peak value at line center is relatively undisturbed. At sufficiently high pressures, the absorption features begin to exhibit significant interference from neighboring transitions. For such asymmetric features, laser intensity modulation may result in $2f$ peak heights that differ from pure $2f$ simulations. However, these higher-order effects may be taken into account in the simulations, as prescribed by equations given in the literature (Liu et al., 2004a; Philippe and Hanson, 1993; Kluczynski and Axner, 1999; Schilt et al., 2003). For a fixed-wavelength $2f$ strategy, pressure-shift effects may cause the laser wavelength to deviate from line-center, especially if large changes in pressure occur such as in a pulsed detonation engine or an internal combustion engine. In such cases, if large modulation depths are employed, it is imperative to simulate the effects of amplitude modulation since they are significant off of line-center, as shown in Figure 3.10.

The simulations shown in Figure 3.10 are performed by numerically integrating (2.15), as described in Section 2.2.1.

![Figure 3.10: $2f$ spectral simulation, adjusted for intensity modulation effects. $i_0 = (2/3)\bar{i}_0$; $a = 0.5$ cm$^{-1}$; $T = 500$ K; $P = 10$ atm.](image)
3.3.2 2f Baseline Calibration

In 2f spectroscopy, the absence of absorption should result in zero signal levels. Unfortunately, for DFB diode lasers, the laser output exhibits a slight nonlinear response to the modulation waveform input. This creates an artificial background 2f signal (in the absence of absorption), a phenomenon that is discussed and analyzed in detail in the literature (Kluczynski and Axner, 1999). In short, when a pure sinusoidal waveform is used to drive the DFB lasers, the nonlinearities distort the pure sinusoid by creating harmonic distortion in the laser intensity modulation. The result of harmonic distortion is that a pure sinusoidal input leads to an output signal with Fourier components at a variety of higher harmonics, including the second harmonic, which looks like a background 2f signal. The magnitude of this distortion signal (background 2f) scales with the amplitude of the modulation. In practice, this background signal may be removed by post-processing. However, for the large modulation depths utilized in this study, the background 2f signal overwhelms the actual absorption-based 2f signal and leads to complications for our lock-in demodulation. Therefore, pre-processing steps are taken instead to eliminate the 2f background signal.

Owing to the digital nature of our waveform generation, it is straightforward to add a second-harmonic component to the pure sinusoidal input waveform generated by the NI-DAQ system. By carefully adjusting the amplitude and phase of this 2f input, relative to the fundamental pure sinusoidal input, it is possible to cancel out the background 2f signal in the laser amplitude modulation. Since the superposition of the input 2f signal on the fundamental driving waveform is linear, there ought to be no noticeable effects on the absorption-based 2f signals. In addition, the input 2f signal amplitudes exploited here are generally only on the order of 1% of the fundamental input sinusoid amplitudes, and should not lead to undesired higher-order effects.

A fast Fourier transform (FFT) of the intensity output of a DFB laser, modulated at 50 kHz, is shown in the following figure, both without (Figure 3.11a) and with (Figure 3.11b) the baseline calibration procedure described above. Note that by introducing a second-harmonic component to the driving injection-current waveform, the power of the
output 2$f$ component is reduced by four orders of magnitude between Figure 3.11a and Figure 3.11b.

Figure 3.11a (upper): Intensity power spectrum for a DFB laser modulated at 50 kHz, WITHOUT baseline calibration and no absorption.

Figure 3.11b (lower): Intensity power spectrum for a DFB laser modulated at 50 kHz, WITH baseline calibration and no absorption.
3.3.3 Noise/Bandwidth Implications on Modulation Depth

Figure 3.12 illustrates how the maximum modulation depth attainable with a typical DFB laser depends upon modulation frequency. Here, the laser is sinusoidally modulated from threshold (about 10 mA) to the maximum current setting of 200 mA ($i_0 \sim \bar{I}_0$). Clearly, lower modulation frequencies enable increased modulation depths. In practice, sensor-bandwidth requirements determine the lower limit to the modulation frequency.

![Figure 3.12: Approximate plot of maximum-attainable modulation depth vs. modulation frequency for a typical laser used in this study.](image)

For the fixed-wavelength WMS sensor developed in this work, the measurement bandwidth of the sensor is determined by the low-pass filter in the lock-in amplifier. Laboratory experiments (Figure 3.13) indicate that a low-pass filter bandwidth of one-third the modulation frequency sufficiently attenuates the fundamental modulation from the $2f$ signal. Figure 3.13 shows that when modulation frequencies exceed a factor of three of the lock-in filter bandwidth, SNR is not improved as there are other noise sources.
which dominate. These noise sources include electronic noise, detector noise, laser intensity noise, and various minor etalons and optical distortions due to optical windows or fibers. For the data shown in Figure 3.13, the rms noise is defined as one standard deviation in the $2f$ signal, acquired with a constant lock-in filter bandwidth of 15 kHz and a 5 MHz sampling rate.

![Figure 3.13: $2f$ noise (rms) vs. modulation frequency with a fixed lock-in filter bandwidth of 15 kHz.](image)

The target measurement bandwidth of this study is 15 kHz. Therefore, the minimum modulation frequency exploited is 50 kHz, resulting in a maximum attainable modulation depth of approximately: $a = 0.8 \text{ cm}^{-1}$.

### 3.4 Validation Facilities

A furnace facility, equipped with a 20 cm path length absorption cell, was used for initial validation experiments of the $2f$ temperature sensor. The optical cell is
designed to withstand pressures up to 10 atm. The tube furnace (Thermolyne F21125) is 40 cm long, of which only the middle 20 cm is occupied by the cell’s path length in order to improve temperature uniformity. Wedged sapphire rods provide optical access and are essential to restrict the path length of the cell to the central 20 cm region. Temperatures of up to 1100 K are attained with pure water samples (1 - 15 Torr), ambient air, or water samples diluted with N₂. Type K thermocouples are attached to the absorption cell at various mid-span locations to monitor the temperature. Maximum nonuniformity in temperature along the path length is on the order of 25 K. When it is desirable to increase absorbance levels, a double pass arrangement is used in order to increase the overall absorption path length to 40 cm. A sketch of the facility is shown in Figure 3.14.

Water vapor mixtures are prepared by filling a small compressed gas cylinder with pure water vapor up to the water vapor pressure at room temperature (about 17 Torr), and then diluting the water with compressed nitrogen up to the requisite pressures. The mixture is mechanically mixed by shaking the gas cylinder, in which Teflon beads are placed to aid in the stirring process. For room-temperature measurements at elevated pressures of up to 20 atm, an unheated static cell with a 100 cm path length is also used.

Figure 3.14: Schematic of the heated optical cell facility
Choosing appropriate spectral features for a multiplexed temperature sensor is not a trivial task. A primary goal is to select spectral features possessing an optimal combination of lower-state energies such that the ratio of recorded signal strengths is a sensitive function of temperature. The theory and strategy behind this optimal selection procedure is discussed in the literature (Arroyo and Hanson, 1993; Zhou et al., 2003). One generally desires strongly-absorbing transitions, although limiting absorption magnitudes is sometimes necessary in certain environments with long path lengths or an overabundance of absorbing species. Also, for the WMS strategies presented in section 2.2, optically-thin conditions are assumed, such that absorption magnitudes should always remain below 10% or so. Depending upon the demultiplexing strategy chosen, one may also need to take the wavelength separation of various spectral features into account. As discussed in the next chapter in Section 4.4, in order to eliminate cross-talk between multiplexed channels, a minimum spectral separation between channels is often required. An additional consideration is the temperature uniformity of the measurement region being probed with path-integrated diode laser techniques. Consider a flat-flame burner with cold edges. Probing a water vapor transition with a small lower-state energy will result in disproportionately large absorption magnitudes due to the cold flame edges. Knowledge of the expected characteristics of a specific measurement application allows one to select spectral transitions that are not misleading and difficult to interpret. These issues have been explored in the literature (Ouyang and Varghese, 1989; Ouyang and Varghese, 1990; Zhou et al., 2003).

For the sensors developed and utilized in this research, certain lines were chosen based on the experience and verification of previous workers (Furlong, 1998; Sanders et al., 2000). More recent work has aimed to identify a relatively strong transition possessing a large lower-state energy for sensitive measurements at elevated temperatures. Strong lines with large lower-state energies are difficult to find, since partition functions increase at higher temperatures as more quantum levels are populated, thereby diluting lower-state populations and reducing all line strengths. Also, spectral
databases such as HITRAN are not as accurate for high-temperature lines, since most previous experiments with NIR water vapor spectroscopy have been performed at low temperatures (Toth, 1994). Therefore, laboratory experiments have been performed in our own heated optical cells in order to validate and improve upon the spectroscopic parameters tabulated in the literature.

Pure water vapor samples (2 - 18 Torr) were prepared and heated in the optical cell facility described in Section 3.4. Experimental line strength results are shown as a function of temperature in Figure 3.15. The heated cell yields data at temperatures up to about 1100 K. A few measurement points at higher temperatures (~1500 K) were obtained by using a 10 cm long flat flame burner fueled with premixed ethylene and air. HITRAN2004 simulations are shown as well, to indicate the discrepancy between measured data and literature values. The high-temperature line at 1335 nm was used in the earliest measurement campaigns (SCRAMJET and gas turbine) before a laser at 1469 nm was obtained. Although HITRAN2004 predicts relatively strong line strengths for the feature at 1469 nm, compared to the feature at 1335 nm, experiments show that this value is overstated by HITRAN2004 and that the feature only exhibits a marginal improvement to the 1335 nm feature. Spectroscopic parameters for the feature at 1335 nm are obtained from HITEMP, the high-temperature extended database for HITRAN.

The results from laboratory validations are listed in Table 3.1. The feature at 1469 nm is an example for which the database-reported line strength (HITRAN2004) is significantly different from measurements. By probing a combination of three water vapor lines in a multiplexed absorption sensor, good temperature sensitivity exists from room temperature up to combustion temperatures of 2000 K or higher. The features at 1392 nm and 1343 nm are used in all measurement campaigns reported here. As mentioned previously, the 1335 nm feature was used in earlier measurements for its high-temperature sensitivity before being replaced by the stronger feature at 1469 nm.
Figure 3.15: Measured and simulated line strengths as a function of temperature for four water vapor features

| Wavelength, nm-HITRAN04 | Wavenumber, cm⁻¹-HITRAN04 | Line strength, $S(296\text{ K})$, cm⁻²atm⁻¹-HITRAN04 | Line strength, $S(296\text{ K})$, cm⁻²atm⁻¹-Measured | Lower-state energy, cm⁻¹-
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<td>1392</td>
<td>7185.60</td>
<td>1.97(10⁻²)</td>
<td>1.96(10⁻²)</td>
<td>1045.1</td>
</tr>
<tr>
<td>1343</td>
<td>7444.35 + 7444.37</td>
<td>1.12(10⁻³)</td>
<td>1.10(10⁻³)</td>
<td>1774.8, 1806.7</td>
</tr>
<tr>
<td></td>
<td>(combined)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1335</td>
<td>7488.71 + 7488.74</td>
<td>1.52(10⁻⁶)</td>
<td>1.5(10⁻⁶)</td>
<td>2739.4, 4438.8</td>
</tr>
<tr>
<td></td>
<td>(combined)</td>
<td></td>
<td></td>
<td>(HITEMP)</td>
</tr>
<tr>
<td>1469</td>
<td>6807.83</td>
<td>1.02(10⁻⁶)</td>
<td>5.8(10⁻⁷)</td>
<td>3319.4</td>
</tr>
</tbody>
</table>

Table 3.1: Line strength and lower-state energy parameters for water vapor features used in measurement campaigns
Chapter 4. Optical Engineering

As the science and spectroscopy of making diode-laser measurements has progressed and proven itself over the years, challenging new applications are now opening up for the implementation of these sensors. The installation of these technologies in realistic combustion devices, such as aircraft gas turbine and SCRAMJET engines, as well as automotive internal combustion engines, presents serious constraints in terms of mounting and optical access. In addition, these turbulent combustion flow fields require carefully optimized optical designs in order to minimize beam steering effects, avoid problems with emission, and enable robust and simple alignment of optical sensor probes.

Much of the following discussion has been the result of efforts to design and implement diode-laser sensor systems for making spatially and temporally resolved temperature measurements in the combustor exit plane of two large-scale propulsion engine test facilities located at Wright Patterson Air Force Base, Ohio (WPAFB). One is a near-atmospheric pressure SCRAMJET test facility and the other is a high-pressure gas turbine sector rig. A single week-long measurement campaign was made at each facility. Many obstacles were encountered and overcome during this development process, with the result being more optimal design concepts that we wish to pass on to the reader. The following discussion provides general design rules intended to guide future work in this area. Specific design choices depend upon the particular engine geometry, environmental conditions, and sensor specifications. Trade-offs are inevitable, as with any design. Readers desiring more detailed information on design choices should refer to Section
3.1.1, Section 3.1.2, and descriptions of individual measurement campaigns found in the next chapter.

Note that only fiber optic-based sensing strategies are considered here. There are many benefits to fiber-coupled designs over free-space beam strategies. The most obvious benefit is the ease of transport of laser radiation since fiber optic cables allow for the convenient routing of laser radiation in and around complicated engine geometries. Beam parameters such as spot size are also independent of path length in a fiber, ensuring scalability of the installation geometry. Fiber optics are also necessary for the purpose of feeding laser radiation through a pressure vessel with minimal intrusion and footprint. It is often unfeasible to machine optical-access windows into high pressure, high temperature combustors. A section of this paper will discuss the design of high-temperature/pressure fiber optic probes for such situations. Many of the latest DFB lasers on the market are manufactured as fiber-coupled modules for the convenience of the telecom industry. This makes a fiber-coupled sensor strategy especially attractive and simple. Most importantly, by combining laser beams in a common single-mode fiber, one may insure that a common free-space path is shared by each laser through the combustor.

4.1 Basic Model

At the center of a diode-laser absorption sensor is the free-space beam path traversed by the laser beam as it interrogates the hot combustion flow field, whether for temperature, concentration, velocity, or some other parameter of interest. A fiber “pitch” takes input laser radiation, collimates the beam, and sends it across a combustor path length onto an opposing “catch” lens which focuses the light into a receiving fiber that, in turn, channels the light to an appropriate detector. A basic diagram model of the components of this pitch/catch system is shown in Figure 4.1.
The inset of Figure 4.1 shows a simplified optical ray trace. An incoming ray of light is generalized to have a radial off-axis distance of $x_1$ and an incident angle of $\alpha_1$ with respect to the normal axis of symmetry. The fiber, the size of which is grossly exaggerated for the sake of clarity, is placed at a distance of one focal length from the lens. The radius of the fiber core is defined as $r$. For a fiber of a specified numerical aperture (NA), $R_{\text{max}}$ is the maximum off-axis distance at the lens for which a ray of light can be successfully coupled into the fiber and transmitted via total internal reflection within the fiber core. Rays of light impinging upon the lens at greater radial distances than $R_{\text{max}}$ are focused onto the fiber at large enough angles such that they transmit across the fiber core-cladding interface instead of being internally reflected and propagated.
If we define a vector,

\[
\begin{pmatrix}
  x \\ \\
  \alpha
\end{pmatrix}
\]  

such that \( x \) is the radial off-axis distance of a ray of light and \( \alpha \) is the angle of incidence (radians), conventional matrix optics can be utilized to analyze the catch system depicted in the inset of Figure 4.1. The following matrix equation relates \( x_1 \) and \( \alpha_1 \) to \( x_2 \) and \( \alpha_2 \):

\[
\begin{pmatrix}
  x_2 \\
  \alpha_2
\end{pmatrix} = \begin{pmatrix}
  1 & f \\
  0 & 1
\end{pmatrix} \begin{pmatrix}
  1 \\
  -1/f
\end{pmatrix} \begin{pmatrix}
  x_1 \\
  \alpha_1
\end{pmatrix}
\]  

(2)

In Equation 2, the 2x2 matrix on the left is the ray matrix for a free-space path of length \( f \). The 2x2 matrix on the right is the ray matrix for a thin lens of focal length \( f \). The result of matrix multiplication is:

\[
\begin{pmatrix}
  x_2 \\
  \alpha_2
\end{pmatrix} = \begin{pmatrix}
  f \cdot \alpha_1 \\
  -x_1/f + \alpha_1
\end{pmatrix}
\]  

(3)

From Equation 3, it is clear that angular offsets in the incident beam, \( \alpha_1 \), produce radial offsets at the fiber, \( x_2 \). Similarly, purely radial offsets in the incident beam, \( x_1 \) (with \( \alpha_1=0 \)), lead to angular offsets at the fiber, \( \alpha_2 \). A nondimensional form of the equations above demonstrates this symmetry if one specifies the nondimensionless variables as follows:

\[
X_1 = \frac{x_1}{R_{\text{max}}} \quad X_2 = \frac{x_2}{r} \quad A_1 = \frac{\alpha_1}{r/f} \quad A_2 = \frac{\alpha_2}{NA} \quad (\text{where } NA = \frac{R_{\text{max}}}{f})
\]
Equation 3 can thus be written in nondimensional form as:

\[ X_2 = A_1 \quad \text{and} \quad A_2 = -X_1 + A_1 \frac{r}{R_{\text{max}}} \]  

(4)

Under the assumption that \( r \ll R_{\text{max}} \), which is true for the optical systems under consideration here, Equation 4 achieves the following form:

\[
\begin{pmatrix}
X_2 \\
A_2
\end{pmatrix} =
\begin{pmatrix}
0 & 1 \\
-1 & 0
\end{pmatrix}
\begin{pmatrix}
X_1 \\
A_1
\end{pmatrix}
\]  

(5)

This is a well-known focal plane characteristic of thin lenses: distances at the lens are mapped into angles at the focal plane whereas angles at the lens are mapped into distances at the focal plane.

### 4.2 Optimization and Constraints.

In almost every post-combustion region of interest, flow-field-induced beam steering effects are a major noise source in laser diagnostic measurements. The beam steering extent is a function of path length, the magnitude of density gradients, and the strength and size of turbulent eddies within the flow field. The geometry of the optical system depicted in Figure 4.1 should be optimized to mitigate the effects of beam steering in order to maximize signal-to-noise ratios (SNR), to reduce averaging time, and to thereby increase sensor bandwidth.

In free-space laser diagnostic systems in which there is windowed optical access to the combustion flow field, one often wishes to maximize detector area in order to compensate for beam steering effects. Unfortunately, it is often necessary to locate the detector as close as possible to the combustion region in order to reduce path length, since beam steering effects are magnified over longer paths. In instances where convenient optical access is unavailable and physical obstructions preclude the placement
of a detector near the combustor itself, fiber optics are invaluable. In such cases, it is the fiber catch component that must be optimized to negate beam steering effects.

From Equations 3 and 5, it is clear that increasing the fiber core diameter, $2r$, and the numerical aperture of the fiber, $NA$, allows the fiber to accommodate angular and radial misalignments, respectively. For short path lengths with highly turbulent flow fields and strong density gradients, angular misalignments may dominate over radial misalignments, requiring larger fiber core diameters to reduce beam steering noise. For long path lengths, radial misalignments tend to dominate, requiring a larger fiber $NA$ and a larger focusing lens to compensate for beam steering.

There are a number of constraints that limit the magnitude of the fiber diameter and $NA$. As the fiber diameter starts to approach the dimensions of the active region of the detector, losses occur due to over-filling of the detector area. As a consequence, mode noise distortion may be introduced from a failure to uniformly couple different fiber modes into the detector. Large fiber diameters of course provide less spatial filtering for the rejection of emission as well.

A few comments should be made about mode noise. The speckled structure of light emitted from multimode fibers is the result of interference between rays of light traveling in different cavity modes within the fiber. Since this structure is wavelength dependent, it can lead to intensity distortions in diode laser wavelength scans if only a portion of the light emitted from a multimode fiber is detected. Mode scramblers or mode mixers are available to mitigate these effects but often suffer from losses and may only be compatible with certain fiber diameters. Single-mode fibers do not exhibit mode noise and are nearly always used as the pitch fiber. However, their small diameters (~9 microns in the near infrared) and small $NA$ (typically 0.1), make them an extremely poor choice for a catch fiber unless there is no beam steering. In order to eliminate fiber mode noise, it is necessary to couple all of the light transmitted by a multimode fiber onto the detector surface. Obviously, if the detector is butted up against the fiber end, this requires a detector with an active area diameter of at least the fiber core diameter. Less obvious is the fact that if the fiber output is collimated and refocused onto a detector, it is impossible to achieve a spot size at the focal point less than the size of the original fiber core (due to etendue conservation). In reality, the detector active area diameter should be
much larger than the fiber core diameter to insure 100% coupling efficiency. Large area silicon and InGaAs detectors in the visible and NIR are available. However, there is a trade-off between detector bandwidth and detector area, as well as detector gain. For example, it is difficult to obtain large-area InGaAs detectors (2 – 3 mm active diameter) with bandwidths of 1 MHz or above. For scanned direct-absorption sensors, this detector speed is sufficient for most sensing needs. However, for a sensor system utilizing WMS, the modulation frequency is typically on the order or 100 kHz, in order to obtain a final sensor bandwidth in the 1-10 kHz range. Therefore, a detector bandwidth of at least 1 MHz is generally necessary. The upshot is that designing a system for optimal beam-steering rejection, through the use of a large multimode catch fiber and a large-area detector, will place constraints on the speed of the diagnostic. Large 3-mm diameter detectors have recently been acquired with 4 MHz bandwidths (Electro-Optical Systems IGA-030-E5/4MHz). The transimpedence gain of these devices is substantially less than for the 10 MHz 1-mm diameter detectors generally used (Thorlabs PDA400).

For multiplexed absorption sensors (see Figure 4.2), it is necessary to separate the various wavelengths collected in the catch fiber. This is often accomplished by using diffraction gratings. Care must be taken to choose a fiber core diameter that allows for effective dispersion of the multiplexed laser wavelengths. The larger the fiber core, the more difficult it is to collimate the output radiation and cleanly disperse the constituent wavelengths with a grating. Cross-talk between channels should be avoided. The divergence angle of a collimated beam from a multimode fiber of core diameter $2r$ is calculated from the upper half of Equation 3: $x_2 = f \alpha_1$. By letting $x_2 = r$, the fiber core radius, we find the divergence angle: $\alpha_1 = r/f \text{ (radians)}$. This formula may be used, along with the dispersion setup path length and the angular dispersion of the grating, to calculate if the demultiplexed beams will spatially overlap on the detectors and lead to cross talk. Typically, a 400-micron diameter, 0.39 NA catch fiber (Thorlabs), along with a 1200 groove/mm grating (Edmund Industrial Optics) are utilized in our demultiplexing setups. In practice, it has been observed that a laser spacing of 10 nm or above is generally sufficient to avoid cross-talk between channels (wavelengths are around 1400 nm). A noteworthy benefit of a grating-based dispersion system is that it allows for the rejection of broadband emission from a combustor. The dispersion optics behave
approximately like a 10-nm band-pass filter to eliminate any flame luminosity captured by the collection fiber. Additional comments on demultiplexing strategies and concerns are made in Section 4.4.

Figure 4.2: Example multiplexed diode-laser absorption sensor installed on a combustor exit plane

Commercially available fibers may be obtained with numerical apertures ranging from less than 0.1 to greater than 0.5, depending upon the core and cladding materials used. The fiber numerical aperture is calculated from the indices of refraction of the core and cladding materials: $NA = (n_{\text{core}}^2 - n_{\text{clad}}^2)^{1/2}$. A typical value for a 200-micron or 400-micron core multimode fiber is 0.40. If the collected laser radiation in the catch fiber must be collimated in free space, as for example in a grating-based dispersion setup, or for emission filtering, large fiber NAs may create difficulties in collimation with standard lenses. To collimate the output of a large NA fiber, a small $f/\#$ lens is required as seen from the standard relation: $f/\# = 1/(2f)$. Because it is difficult to find lenses with $f$-numbers much less than unity, the choice of fiber NA is limited in certain cases. It is often beneficial to use aspheric lenses to avoid spherical aberrations and other non-thin-lens effects at the periphery of the lens. Unfortunately, these lenses are generally
manufactured using an industry-standard molding process using low-melting-point lens materials. Therefore, they are not appropriate for utilization in high temperature fiber optic pitch and catch probes.

Finally, in most realistic combustion devices, emission levels are high and may obscure laser signals. Band-pass filters may be used to filter out broadband emission noise. In grating-dispersed multiplexed sensor systems, the grating itself acts as a filter. Gratings with greater dispersive power narrow the effective filtering band pass. If these methods all fail to reduce emission levels to acceptable levels, one must then reduce the fiber NA or fiber diameter in order to spatially filter the emission. The coupling efficiency of emission into a fiber catch scales linearly with fiber NA and quadratically with fiber core diameter. Therefore, reducing the fiber core size provides the greatest reduction in emission levels.

### 4.3 High Pressure and Temperature Fiber Optic Probes.

As laser diagnostic technologies mature, they are finding applications in more realistic combustors. Unfortunately, this often comes at the price of severe spatial constraints and optical access limitations. The availability of both single-mode and multi-mode fibers capable of withstanding high temperatures has enabled the development of high temperature and pressure fiber optic probes. Generally, these high-temperature fibers are jacketed with metals such as gold or copper. This allows them to be swaged or welded into hermetic probe designs without the use of low-temperature epoxies or glues. In our probes, the fibers are protectively housed in a rigid 1/8” stainless steel tube that is welded to a ¼” probe tip, also constructed entirely of stainless steel and holding a sapphire lens. The probe tip contains a lens for collimation in pitch probes or for the focusing of received light in collection probes. The longitudinal lens position is adjusted via two threaded lens-holder rings, again avoiding low-temperature adhesives. A sketch of the high-temperature, high-pressure probe (Fiberguide Industries, custom design) used in gas turbine measurements is shown in Figure 4.3. The rigid 1/8” stainless steel tube may be replaced by a flexible stainless steel monocoil design if needed.
4.4 Demultiplexing Strategies

Diode laser ratio thermometry is generally performed by multiplexing several laser light sources in order to probe spectroscopic transitions at various wavelengths. Single-laser ratio thermometry with water vapor in the NIR is possible (Xin et al., 2004) but is heavily constrained by the need to find spectrally-adjacent water features that are sufficiently strong absorbers and possess different temperature dependencies. Depending upon the specific measurement application, this is often difficult to do, especially since adjacent water features often have similar lower-state energies that cause them to behave similarly with temperature (not ideal for thermometry).

In this work, the multiplexing of fiber-coupled DFB lasers is accomplished through the use of standard single-mode fiber combiners. Custom grating-based multiplexers are a possibility, and would be much more efficient, but have not been pursued yet. Several demultiplexing strategies are possible, and possess individual strengths and weaknesses. The following discussion will explore some of those trade-offs. Three categories will be covered here: wavelength, modulation-frequency, and time-based demultiplexing.

Figure 4.3: High-temperature, high-pressure probe used for gas-turbine sector rig diagnostics.
4.4.1 Wavelength Demultiplexing

Wavelength-based demultiplexing may be accomplished through the use of a dispersive medium, such as a prism or a grating, or through the use of spectral filters. In most cases, avoiding cross-talk between channels requires a relatively large spectral separation between the multiplexed light sources. For the grating-based system described in Chapter 3, a spectral separation of 10 nm between channels is necessary to avoid cross-talk. This separation could be reduced through the use of a smaller collection fiber than the 400-micron fiber used here, since laser beam collimation is improved with smaller fibers. The divergence half-angle of a collimated beam from a multimode fiber may be expressed as $r/f$ where $r$ is the fiber core radius and $f$ is the focal length of the collimating lens. Increasing the focal length clearly reduces the divergence angle, but at the expense of a larger beam diameter, given by $2f(NA)$, where NA is the fiber numerical aperture. Custom gratings based on higher diffraction orders may also be obtained for improved dispersion. Such echelle gratings are often manufactured with high efficiencies and may be optimal for certain dispersion requirements, but have not been explored in this work.

Diffraction gratings are a mature technology and have been the method of choice for the majority of the demultiplexing needs of this work. Figure 4.4 is a schematic of this strategy utilized in a multiplexed gas sensor. The one drawback of this strategy is the bulky hardware, which includes separate detectors for each channel, focusing mirrors, the grating, and collimating optics, all aligned in a tent or enclosure designed to purge ambient water vapor. An alternative to this method is to use interference filters, either in a free-space configuration, or fiber coupled. Band-pass filters may be purchased for specific wavelengths, but do not provide the flexibility that gratings do for experimenting with different laser wavelengths, which is beneficial for the development work done here. Once again, filter response curves must be taken into account, along with laser wavelength selection, in order to eliminate cross talk between channels.
A few comments should be made about purging the dispersion setup. This is necessary because the species detected in this work, water vapor, is present in ambient air, which causes a background interference signal. The free-space path length in these dispersion setups is often much longer than the measurement path lengths. The need to purge depends on the lower state energy, $E''$, of the water vapor absorption lines chosen for the sensor. Lines with small lower state energies are strongest at room temperatures and require more aggressive purging to eliminate ambient water vapor interference. For a high-temperature sensor incorporating lines with high lower state energies, rejecting room-temperature ambient water vapor may not be as much of an issue.

### 4.4.2 Modulation-Frequency Demultiplexing

If modulation spectroscopy is used, there is the potential to demultiplex laser signals by utilizing different modulation frequencies for each source, similar to how an FM radio operates to separate individual channels. The benefits of this method are that laser sources are not required to be spectrally separated at all. In addition, only one detector is required, resulting in a much-simplified optical setup and alignment. That
single detector signal is analyzed with lock-in amplifiers tuned to perform phase-sensitive detection at a specific modulation frequency or harmonic component. The drawback of this method is that in order to eliminate crosstalk between channels, it is necessary not only to modulate the lasers at distinctly different frequencies, but one must choose the modulation frequencies carefully to avoid aliasing and harmonic interferences. The result of these constraints on modulation frequency is to constrain the sensor bandwidth and the modulation depth of the lasers, as discussed in Section 3.3.3. For this reason, it is somewhat impractical to implement this strategy for more than two channels in a high-speed sensor: separating the modulation frequencies sufficiently to avoid cross-talk would impose serious limitations on modulation depths and sensor bandwidth. Figure 4.5 is a schematic of this strategy.

![Figure 4.5: Modulation-frequency-based multiplexing and demultiplexing.](image)

From Figure 3.11a, it is apparent that a signal modulated at $1f$ will possess harmonic components at $2f$, $3f$, and so on. The low-pass filter bandwidth determines the frequency range over which the lock-in amplifier acts as a band-pass filter to extract signals at specified frequencies. For example, if a 15 kHz lock-in filter bandwidth is utilized for $2f$ detection of one channel, one must insure that the other channel does not contain harmonic components within at least 15 kHz of the detected $2f$ frequency. Since no filter is perfect, a 30 kHz separation will more-realistically be required, given a 15
kHz filter bandwidth (-3 dB bandwidth). As an example, consider two channels modulated at fundamental frequencies of $f_1 = 70$ kHz and $f_2 = 87.5$ kHz. The separation between $2f$ frequencies is: $2f_2 - 2f_1 = 35$ kHz. One must also verify that the third harmonic of $f_1$ is sufficiently isolated from $2f_2$. Indeed, $3f_1 - 2f_2 = 35$ kHz as well. Note that in order to achieve these 35 kHz separations, it is necessary to choose modulation frequencies that are both significantly higher than the minimum modulation depth required for a single lock-in output at 15 kHz, which was previously shown to be about 50 kHz (see Figure 3.13). This requirement for higher modulation frequencies has a detrimental effect on the maximum-attainable modulation depth (Figure 3.12).

### 4.4.3 Time-Division Multiplexing

Time-division multiplexing is achieved by alternating laser scans using different sources, switching back and forth in time between channels. This allows for simple optical setups, with single detectors, but degrades sensor bandwidth due to the obvious reason that each channel is operational for only a fraction of the time. In addition, information is gathered periodically rather than continuously, with “dead-time” between readings. For these reasons, time-division multiplexing is rarely used, but in situations where bandwidth is not of major concern, this strategy may be useful and relatively straightforward to implement.
Chapter 5. Measurement Campaigns

Various measurement campaigns have been made using multiplexed diode-laser sensors probing water vapor features in the near-infrared. Depending upon the specific application, both fixed-wavelength and scanned-wavelength direct-absorption strategies have been employed in these practical test facilities.

Advances in combustion engines are being enabled by modern technologies that improve upon the capabilities and efficiencies of current engines. Improvements come in the form of pollution reduction, better fuel economy, and increased engine output. Many of these improvements rely upon advanced sensing and control strategies to insure that optimal engine conditions are attained. For example, engine “pattern factor” refers to temperature profiles and fluctuations in the post-combustion region of a propulsion device. The sensors developed in this work are a much-needed component in modern efforts to control and improve propulsion engine pattern factors. The objective of this research is to develop and implement diode-laser-based absorption sensors to monitor key combustion parameters, such as temperature and water vapor concentration, in practical aircraft engines.

5.1 SCRAMJET Test (September, 2002)

A multiplexed diode-laser sensor system was installed in the post-combustion zone of a SCRAMJET engine test rig located at Wright Patterson Air Force Base. The
water-based, scanned-wavelength absorption sensor was used to determine path-integrated temperature and water vapor mole fraction across the test region.

Figure 5.1 is a schematic of the SCRAMJET facility. An electrically-heated dry air flow is burned in a vitiator, which heats the flow and raises its pressure so that it can be expanded to supersonic speeds. The supersonic flow, at about 1 atm, is burned in the main combustor, after which an optical access window is located. The rectangular window is oriented vertically and is about 1” by 6” in dimension. Engine runs could be sustained for about one minute, during which the sensor collected data for a few seconds at a time at sampling rates of 1 to 5 MHz.

![Figure 5.1: Schematic of the SCRAMJET test facility at Wright-Patterson Air Force Base, Ohio.](image)

The multiplexed sensor utilized three fiber-coupled diode lasers at 1335, 1343, and 1392 nm (see Section 3.5 for details), each of which interrogates absorption from a selected water vapor transition. The ratios of the absorption strengths of the features were used to infer gas temperature, and the absorption in a single transition then yielded water mole fraction. The three beams were multiplexed into a single fiber using a 3x1 fiber-combiner. The multiplexed beam was sent through the test region, collected into a 50-µm fiber for this preliminary test, and demultiplexed onto three separate detectors using a grating-based dispersion system. Two different scan rates were employed: 1 kHz
and 5 kHz. A description and schematic example of the sensor instrumentation were provided in Section 3.1.1.

Due to interference from the Fabry-Perot etalon created by internal reflection within the 1”-thick quartz windows, it was necessary to angle the laser beams paths through the windows by a significant amount (~10 degrees) as shown in Figure 5.2. Anti-reflection coatings are not feasible due to the high temperatures involved, which would vaporize window coatings. Wedged windows, to eliminate etalons, have been designed for future tests.

![Diagram of alignment through the SCRAMJET combustor optical access windows](image)

Figure 5.2: Alignment through the SCRAMJET combustor optical access windows

Nitrogen purge tubes and bags were installed to eliminate atmospheric water interference in the sensors. By analyzing the absorption levels with a pre-heated dry air flow turned on in the SCRAMJET rig, it was verified that the purge lines eliminated most, if not all, of the ambient water vapor along the beam paths. In addition, by scanning the lasers from below threshold, so that the lasers were off during a portion of each scan, it was possible to ascertain that there was no flame emission noise on the detectors. Good rejection of flame emission is due to the spatial filtering provided by the fiber-optic collection system, as well as the spectral filtering afforded by the demultiplexing setup. The dispersion of the diffraction grating (1200 lines/mm, 1st order) in the demultiplexer acts approximately as a 10 nm bandpass filter for each channel.
Details of the experimental setup are given in Section 3.1.1. No additional filters were required to reject emission.

Figures 5.3 and 5.4 are photographs of the SCRAMJET facility. The figures demonstrate the need for fiber optics due to the physical constraints of the system.

Figure 5.3: View of the SCRAMJET test facility at WPAFB, OH
In SCRAMJET measurements, signal quality suffered as a result of turbulent flow field-induced beam-steering noise. The single laser scan shown in Figure 5.5 demonstrates the extent of the beam-steering noise. Figure 5.6 shows the same measurement after a 100-scan average is performed. For the combustor run shown in Figures 5.5 and 5.6, a ratio of integrated absorbance (line strength) of the water vapor features at 1392 and 1343 nm yields an average temperature of about 1200 K. The measured water mole fraction is about 10%. The low temperature reading indicates a combustor problem, which is consistent with the fact that the combustor completely failed several runs later that evening.
Figure 5.5: Single laser scan at 1392 nm in a SCRAMJET test facility. $T \sim 1200$ K, $P \sim 0.85$ atm, $L = 25$ cm, $\chi_{\text{water}} \sim 10\%$.

Figure 5.6: 100-scan average at 1392 nm in a SCRAMJET test facility. $T \sim 1300$ K, $P \sim 0.85$ atm, $L = 25$ cm, $\chi_{\text{water}} \sim 10\%$. 
Since these data were taken, the beam-steering noise problem has been addressed through optical design modifications. For example, a 400-micron collection fiber has replaced the 50-micron fiber. The 8x improvement in fiber diameter, as well as the 2x improvement in fiber numerical aperture, has proven effective in addressing beam steering problems. In addition, collection lenses with larger inlet apertures (Thorlabs F220SMA-C) have been acquired to optimize the fiber optic catch hardware.

The evidence from our preliminary analysis indicated that there were significant spatial non-uniformities in the SCRAMJET flow field. Future measurements goals are to verify and quantify this non-uniformity by probing multiple locations, utilizing vertical translation stages on either side of the optical access window, with improved optical hardware.

### 5.2 Gas Turbine Sector Rig (November, 2002)

Lasers at 1392, 1343 and 1335 nm (see Section 3.5 for details) were used to probe high-pressure water features in a gas turbine sector rig located at WPAFB, Ohio. The gas turbine combustor section operates at pressures of up to several hundred psi, and is located within a larger pressure vessel under the same pressure condition. Temperatures within the combustor are around 2000 K, while temperatures in the air flow surrounding the combustor (but within the pressure vessel) may reach 750 K. A description and schematic example of the sensor instrumentation were provided in Section 3.1.1.

The high pressure and temperature environment of the gas turbine sector rig placed severe access limitations on optical diagnostics. These access problems were solved using fiber optics to transport the light to (transmitter) and from (receiver) the combustor exit plane, shown schematically in Figures 5.7 and 5.11 and shown photographically in Figure 5.8 (surrounding pressure vessel). Engine runs could be sustained for several minutes, during which the sensor collected data for a few seconds at a time at sampling rates of 1 to 5 MHz.
Figure 5.7: Gas turbine sector rig optical access.

Figure 5.8: Gas turbine sector rig pressure vessel at WPAFB, OH
Custom high temperature fibers optics (Fiberguide Industries), with specialty metallic coatings, were fabricated into stainless steel probes that were fed through the pressure vessel wall and into a “window frame” mount (Figure 5.9) surrounding the post-combustion gases at the exit plane of the combustor. Details of this design were provided in Section 4.3. Each ¼” diameter transmitter probe tip contained optics to collimate and pitch the laser light across the 3” width of the combustor exit plane. The transmitted light was collected into a receiver fiber with optimized optical components mounted in similar hardware.

Water-cooling and air-cooling channels in the mounting hardware were successful in keeping the operating temperatures below the 1000 K limits of the fiber optics. A nitrogen purge flow channel around each probe tip was also designed to cool the lens surface and prevent fouling of the optics. The fabricated probe tips were only ¼” in diameter and optical access required only 3/16” diameter penetrations in the combustor wall. In the test rig these probes were inserted into the “window frame” mount at the combustor exit plane (Figures 5.9 and 5.11).

Figure 5.9: “Window frame” mount at the Pratt and Whitney gas turbine sector rig, WPAFB, OH
Each transmitter (pitch) fiber probe consisted of a multi-mode, 50-micron core, high-temperature fiber mated to a sapphire exit lens. Two receiver fiber probe designs were tested, one identical to the 50-micron transmitter probe, and the second probe design utilizing a 400-micron core, high-temperature fiber. The 400-micron fiber probe also had a numerical aperture twice as large as the 50-micron fiber probe, and offered a significant performance advantage. Only the 400-micron collection probe provided reasonable signal strengths, and was used exclusively for data collection. A description of the probe design was provided in Section 4.3, with a schematic shown in Figure 4.3.

An example of absorbance data obtained from the sector rig is shown in Figure 5.10. The ratio of the peak absorbance of the two lines is directly related to gas temperature as shown on the right side of Figure 5.10.

The absorption data collected during these tests are the first-ever measurements of diode-laser absorption in a gas turbine combustor. The example presented in Figure 5.10 extracts a temperature of 2650 °F. This data clearly shows the feasibility of using a
diode-laser sensor for real-time temperature measurements in a gas turbine combustor. These data were averaged over 200 scans, which corresponded to 0.2 seconds of continuous sampling. Temperature measurements along three different measurement paths are shown in Figure 5.11 for one measurement condition at $p = 100$ psi. The measurements indicate significant spatial variations in temperature, perhaps correlated to the combustor fuel injector locations. The temperatures were obtained by averaging a second of data (1000 scans taken at 1 kHz).

![Combustor “window frame” optical mount and preliminary measured temperatures for one run condition at $P = 100$ psi.](image)

Although measurements with the multimode transmitter provide proof of concept validation of our gas temperature measurement strategy, the quantitative temperature data is distorted by mode noise from the multimode transmitter fiber (see Section 4.2). This noise seriously reduces the ability to obtain accurate baseline fits because it introduces distortion in the laser ramp signal. In addition, the absorption feature itself is distorted, increasing the error. It is believed that without the mode noise, the analysis of our data
would be much more robust and would lead to consistent results. In future work, the multimode transmitter fiber will be replaced with a single-mode transmitter fiber design.

Maintaining beam alignment within the gas turbine combustor window frame mount is also a problem, particularly due to mechanical and thermal stresses introduced when the engine is operating. For example, during combustor runs, the engine thrust causes the combustor section to shift nearly one inch with respect to the external pressure vessel. Since the fiber optic probes are rigid and penetrate both the pressure vessel and combustor walls, this creates enormous stresses on the probe, leading to misalignments. Mechanical improvements are therefore necessary in the mounting of the high temperature fiber optic probes in future tests. The challenges are compounded by spatial constraints, high pressures, high temperatures, and run-time transients (mechanical and thermal).

5.3 Pulsed Detonation Engine Rig (December, 2003)

A water vapor sensor was transported to China Lake Naval Weapons Center and installed on one of the exit tubes on a Pratt and Whitney five-tube pulsed detonation engine (PDE) test facility. Three water vapor transitions were interrogated with DFB lasers at 1392 nm, 1343 nm, and 1469 nm (see Section 3.5 for details) in order to obtain temperature information in the engine. A non-resonant beam at 1315 nm was also multiplexed with the resonant lasers in order to provide information on beam-steering and vibration noise. The non-resonant signal corrects for common mode noise. By taking a ratio with the non-resonant beam, SNR is improved by about a factor of three. Extremely strong vibrations were an important noise component in these measurements. The PDE tubes were operated with nozzles for these tests, increasing the magnitude of the vibrations. Previous measurements on a PDE facility at the Naval Postgraduate School in Monterey, CA, have been published and provide additional information and references describing this new engine technology (Sanders, et al., 2000).
Figure 5.12 is a photograph of the experimental facility at China Lake where the Pratt and Whitney PDE was located. Most of the equipment, including the lasers, detectors, dispersion optics, and DAQ system, were housed in a remote electronics bunker, of which a portion of the interior is shown in Figure 5.13. A long single mode fiber provided signal to the PDE and a long 400-micron multimode fiber returned the collected signal to the bunker for demultiplexing and detection. All personnel were located in a concrete control building about 50 yards from the engine facility. An ethernet cable buried underground connected a remote desktop computer with the DAQ computer located in the electronics bunker.

Figure 5.14 is a photograph of the PDE itself, which includes five cylindrical detonation tubes circularly arranged (Kelly, 2003). Optical diagnostics were performed on only the top tube, where threaded ports allowed optical mounting flanges to be screwed into place, as shown in the schematic in Figure 5.15. The mounting flanges all contained a thick wedged sapphire window allowing optical access into the detonation tube. For the pitch side, a collimating unit (Thorlabs F230FC-C) was used to send laser radiation from a single mode fiber through the detonation tube. For the collection side, an aluminum “L-bracket” attached to the optical flange (Figure 5.15) held a collimating unit (Thorlabs F220SMA-C) and a 400-micron catch fiber mounted in a 5-axis stage (Newport LP-05A). The PDE tubes are fired sequentially at a high repetition rate for one to two seconds at a time. The water vapor temperature sensor collects data for the duration of the one to two-second firing series, which consists of many detonation events.

The PDE operates with a vitiated fuel/air mixture. The buffer air is generally heated to a temperature of about 500 K before the engine is run. The main detonation tubes, shown in Figure 5.14, and schematically in Figure 5.16, are ignited with the aid of smaller initiator detonation tubes located deep within the main tubes (Figure 5.16). The first few detonation events of each run are predetonations in which only the initiator tubes are filled with fuel/air and are fired. Compared to the firing of the main tube, engine vibrations are substantially reduced during these predetonation-only firings. Figure 5.16 shows the locations of ethylene fuel sensors as well, which were installed by another student.
Figure 5.12: Pratt and Whitney pulsed detonation engine located at a test range at China Lake Naval Weapons Center, California

Electronics bunker (lasers, detectors, optics, DAQ computer)  Pulsed Detonation Engine

Figure 5.13: Data acquisition and laser equipment for the China Lake PDE sensor
Figure 5.14: Pratt and Whitney multi-tube high repetition rate pulsed detonation engine located at China Lake, CA

Figure 5.15: Optical mounting schematic on the PDE tube
Both fixed-wavelength and scanned-wavelength direct absorption strategies were used in these PDE measurements. A description and schematic example of the sensor instrumentation were provided in Sections 3.1.1 (scanned-wavelength) and Section 3.2.1 (fixed-wavelength). The wavelength scans at 1 kHz provided absolute absorbance information during the engine cycles by sweeping the laser across entire absorption line shapes. Except for the first few milliseconds after the detonation spike in each engine cycle, the total pressure within the PDE tubes was below 5 atm and generally reached a plateau value of about 2–3 atm during the blow down process and the buffer air/fuel fill. Therefore, absorption line shapes were relatively isolated and distinct for most of the engine cycle, and integrated absorbance ratios were assumed to scale directly with line strength ratios to infer temperature. While the scanned absorption strategy yielded absolute absorbance measurements at 1 kHz, a fixed wavelength operation was used to increase measurement bandwidths to the data-acquisition sampling rate of 1 MHz. Post-filtering of the data reduced the measurement bandwidth to about 50 kHz, which was sufficient to resolve temperature fluctuations in this high-repetition-rate PDE facility. The fixed-wavelength sensor was calibrated for each cycle by assuming a buffer/fuel fill temperature of 500 K. This is the target temperature of the buffer air and fuel mixture that is injected into the PDE tubes prior to every detonation event. Independent
temperature measurements from the scanned-wavelength sensor show that the 500 K buffer/fuel temperature assumption is accurate.

The results from this measurement campaign demonstrate the ability of our diode-laser sensor to resolve detonation cycle characteristics and time scales. Figure 5.17 shows the detected laser intensity at 1343 nm for a single PDE cycle. The laser intensity clearly decreases after the detonation occurs due to an increase in combustion-product water vapor, which absorbs the laser radiation at 1343 nm. As the gas temperature cools following the detonation spike (from about 3000 K down to 2000 K), the laser intensity drops further as absorption increases, since the line strength of the 1343 nm feature peaks at around 900 K. Also, total pressure decreases during the post-detonation blow down period, which acts to narrow the absorption features and increase peak absorption magnitudes. The arrival of buffer-air purge dilutes the water vapor concentration, which reduces absorption and causes the laser intensity to increase once again. The fuel arrival provides additional dilution and cools the mixture back to 500 K, in anticipation of the next detonation event.

Figure 5.17: Transmitted laser intensity at 1343 nm (7444.36 cm\(^{-1}\)) for a single PDE cycle

Figure 5.18 shows an example of two engine cycles, presented in absorbance units in contrast to the transmitted intensity shown in Figure 5.17. The second cycle is an
example of an engine misfire. Pre-ignition of the fuel/air mixture is visible, since the water vapor concentration and temperature remains high, resulting in high absorbance levels in the second cycle shown.

By measuring the time between detonation and the arrival of buffer air for a series of detonations, it is possible to monitor buffer timing fluctuations, as shown in Figure 5.19. Much of the scatter in Figure 5.19 may be due to sensor noise, but it is clear that the first few detonations exhibit a larger amount of scatter that settles over time to a nominal value of about ±2%.

Figure 5.18: Example of a healthy engine cycle (left) and a misfire (right). Absorbance plots are at 1343 nm.
In order to determine temperature, the ratio of peak absorbance values from two water features is assumed to vary directly with line strength ratio (to rough approximation). Figures 5.20 and 5.21 show temperature measurements for a series of predetonation events in which only the initiator (ignitor) tube in the PDE is firing:

Figure 5.20: “Run 746”; predetonations #5-7
The results demonstrate the cycle to cycle reproducibility of the temperature measurements. The large noise spikes at the highest temperatures are due to vibrations (about 1 kHz frequency) coupled with the fact that absorption line strengths are weaker at higher temperatures, leading to greater error levels. The peak temperatures measured in Figure 5.21, for “run 747,” are noticeably higher than the corresponding temperatures shown in Figure 5.20, for “run 746.” This is consistent with an increased fuel fill utilized for the later run.

An example of a temperature measurement for one full detonation cycle is shown in Figure 5.22. Once again, this temperature plot is constructed by assuming that absorbance ratios scale directly with line strength ratios, which is true if the spectroscopic features broaden similarly with pressure and temperature, such that line shape changes are identical for each feature. This is not a completely accurate assumption in this case, especially since the 1343 nm feature is actually comprised of two closely-separated transitions, but the approximation is certainly a good one considering the overwhelming effects of vibration and beam-steering noise affecting the signals.
Figure 5.22: Temperature measurement for a full detonation cycle.
Chapter 6. WMS Laboratory Validations

6.1 Low-Pressure 2f Thermometry

$2f$ validation experiments were conducted in a heated optical cell, as described in Section 3.4. The theory and optimization of $2f$ peak ratio thermometry, explored experimentally here, was discussed previously in Section 2.2. Experimental details were described in Section 3.1.1. An example plot of $2f$ peak heights, adjusted to account for laser transmission fluctuations as discussed in Section 3.1.2, versus temperatures from 450 K to 1075 K, is plotted in Figure 6.1 for the absorption feature at 1392 nm. The absorption cell is filled with various amounts of pure water vapor ranging from 1.5 Torr to 2.5 Torr, all of which result in peak absorbance levels of less than 10% (satisfying Equation 2.8). Three modulation amplitudes are used at each condition near the maximum modulation index: $m \sim 2.2$. Note that with increasing temperature, the value of the modulation amplitude that yields a maximum $2f$ peak height changes slightly. As discussed previously, this is due to an increase in line width, which is predominantly Doppler-broadened in this case, as temperature rises. The Doppler half-widths of the water vapor samples increase from .013 cm$^{-1}$ to .02 cm$^{-1}$ over the measured temperature range, and the collision half-widths decrease from .0005 to .00025 cm$^{-1}$, according to the spectroscopic parameters listed in the HITRAN2000 database. As line width increases, so does the value of modulation amplitude, $a$, needed to achieve the maximum $2f$ signal at $m \sim 2.2$ (see Equation 2.10 and Fig. 2.1). As shown in Figure 6.1, the modulation depths used in these measurements range from about .030 to .061 cm$^{-1}$, corresponding to
modulation indices, \( m \), ranging from approximately 2.3 to 4.7 at 450 K and 1.5 to 3.0 at 1050 K. Note that the general shape of the curve in Figure 6.1 reflects the shape of the analogous line strength vs. temperature plot shown in Figure 6.2. The success of our temperature sensor relies on the fact that these two quantities, the line strength and \( 2f \) peak height, are equivalent functions of temperature for appropriate choices of \( m \). The two features plotted in Figure 6.2, at 1392 and 1343 nm, are the two features used in the low-pressure \( 2f \) thermometry validation experiments described below.

![Graph of adjusted \( 2f \) peak height versus temperature.](image)

**Figure 6.1:** Adjusted \( 2f \) peak height versus temperature. 1392 nm feature at three modulation depths. See text for details.
Second harmonic peak ratios, at the measurement conditions stated above, are plotted for the 1392 nm and 1343 nm features in Figure 6.3 and Figure 6.4. Figure 6.3 covers the entire temperature range of the furnace measurements and is normalized such that the largest ratio is unity. Figure 6.4 covers a smaller temperature span at the higher temperatures, and is again normalized using the largest ratio in the plot. Note that although various modulation depths are used in Figure 6.4, corresponding to modulation indices ranging from about 1.5 to 3.5, the use of peak ratios causes the results to collapse onto one curve. This demonstrates one of the simplifying effects of matching the modulation indices for each line. Line strengths and lower state energies from the HITRAN2000 database are used to calculate line strength ratios, which are also normalized and plotted in the same Figures. Considering the effects of temperature uncertainty and nonuniformity in the cell (~25K), the data matches the simulated $2f$ ratios (based on line strength ratio) well. The agreement between $2f$ peak ratios and line strength behavior, as a function of temperature, demonstrates an additional simplifying effect of optimizing modulation depths (see Section 2.2.2).

Figure 6.2: Simulated line strengths versus temperature (HITRAN2000 database)
Figure 6.3: Normalized $2f$ peak ratios versus temperature. 1392 nm signal / 1343 nm signal. Simulations utilize HITRAN2000.

Figure 6.4: Normalized $2f$ peak ratios versus temperature (reduced temperature range). 1392 nm signal / 1343 nm signal. Simulations utilize HITRAN2000.
The need exists to monitor real-time temperature fluctuations in pre and post-combustion flow fields of realistic combustion devices. The temperatures in such flow fields can vary by hundreds of degrees (Kelvin). Figures 6.3 and 6.4 clearly demonstrate the potential of a multiplexed 2\(f\) diode-laser sensor for rapidly and accurately monitoring temperatures in gaseous flows (1 kHz bandwidth in this case). For applications in which a fixed-wavelength implementation of 2\(f\) spectroscopy is feasible, higher measurement bandwidths may be attained, limited only by the lock-in time constant (50 kHz in this case). For fixed-wavelength 2\(f\) peak height measurements, transmission fluctuations must not exist, or if they do, absorbance levels must be low such that \(I_t\) may be assumed to be identical with \(I_0\). The data in Figure 6.4 can be presented in an alternative form to demonstrate the effectiveness of the sensor to infer temperature. In Figure 6.5, the data point at 825 K, with \(m \sim 2.2\), is used to calibrate the 2\(f\) sensor. Temperatures at four hotter conditions are extrapolated by using the calibration point and the HITRAN2000-simulated temperature dependence of the line strength ratios. The agreement of the 2\(f\) sensor measurements with the thermocouple measurements is within the uncertainty and nonuniformity of the thermocouple and furnace (±25 K).

![Figure 6.5: Calibrated 2\(f\) temperature measurement versus thermocouple measurements (using 1392 nm / 1343 nm peak ratios). \(P = 1.5 – 2.5\) Torr.](image-url)
A comparison of the SNR of direct absorption measurements with WMS measurements shows a significant improvement of nearly one order of magnitude for 2f line shapes (~0.02% minimum-detectable absorption for a 1 kHz bandwidth). This is consistent with what is found in the literature [Bomse, et al., 1992; Hovde and Parsons, 1997]. Also, there is no ambiguity in the baseline of the 2f line shape, which eliminates the baseline-fitting errors common with direct absorption measurements, resulting in more reproducible measurements. This is especially true for weak absorption or if neighboring water lines interfere with the spectral feature and obscure the zero-absorption baseline, which is the case for most lines at higher temperatures and pressures. In certain cases, neighboring features may be broadened and are close enough to lift the baseline trough to a finite absorption level, eliminating the zero-absorption baselines entirely. 2f signals are sensitive to line shape curvature (the 2nd derivative), making them insensitive to baseline. Second harmonic line shapes are determined by instrument settings and lock-in parameters, which are reproducible and eliminate the need for manual post-processing. The convenience in data processing, along with an improved SNR, gives the 2f sensor major advantages for accurate and fast temperature monitoring in many applications.

Finally, in order to generalize our experimental validations to atmospheric pressure situations, identical temperature measurements were carried out on ambient water vapor at atmospheric pressures in the furnace facility. At these conditions, the water absorption features may be modeled as Voigt line shapes that are more heavily pressure-broadened than Doppler-broadened. Figure 6.6 shows a temperature measurement analogous to the low-pressure plot of Figure 6.5. According to the HITRAN2000 database, collision half widths of the air-broadened ambient water vapor sample are about 0.053 cm\(^{-1}\) and Doppler half widths are about 0.011 cm\(^{-1}\) at the measured conditions. A modulation depth of about 0.14 cm\(^{-1}\) was used for the measurement shown in Figure 6.6, corresponding to a modulation index, \(m\), near 2.2. The excellent agreement with thermocouple measurements demonstrates the utility of the 2f ratio technique for making temperature measurements in a variety of pressure conditions up to atmospheric.
6.2 High-Pressure $2f$ Thermometry

High-pressure $2f$ validation experiments were conducted in the heated optical cell (up to 10 atm) as well as the room-temperature cell (up to 20 atm) described in Section 3.4. A theoretical description of high-pressure $2f$ spectroscopy for temperature and species was given in Section 2.2.3 and 2.2.4. Experimental details and strategies for achieving high modulations depths were covered in Sections 3.2 and 3.3. Since $2f$ signals are a function of pressure and temperature, separate experiments were devised to study $2f$ signal behavior as a function of each of those parameters.

6.2.1 $2f$ versus Pressure

A meter-long gas cell at room temperature was used to record $2f$ peak heights of a water vapor transition at 1371 nm at a variety of pressures from 5 atm to 20 atm. $2f$
simulations based on the HITRAN2004 spectral database were calculated and compared with laboratory measurements. The room-temperature results in Figure 6.7 are of $2f$ signal strengths versus pressure at two modulation depths: $a = 0.5 \text{ cm}^{-1}$ and $a = 0.65 \text{ cm}^{-1}$. A one-point calibration is used to scale the simulations to the $2f$ signal units. Note that a separate calibration is not required for both modulation depths, as the $2f$ simulations account for the magnitude of $a$. Figure 6.8 is a simulation of the absorption line shape of the water feature used in this experiment at three pressures: 5 atm, 10 atm and 20 atm. The modulation frequency used was 50 kHz, with a lock-in filter bandwidth of 15 kHz.

![Graph](image)

Figure 6.7: $2f$ peak height versus total pressure. 1371 nm (7294.1 cm$^{-1}$) water vapor feature. $T = 296 \text{ K}$. Simulations utilize HITRAN2004.

Discrepancies between data and simulation (Figure 6.7) may be attributed to errors in HITRAN, in which the broadening parameters are especially suspect. The water vapor mixtures used in these experiments are dilute, since the vapor pressure of water, about 17 Torr, is mixed with over 20 atm (15,200 Torr) of nitrogen, resulting in a water vapor mole fraction of about 0.001.
From Figure 6.7 (and Figure 1.2), it is apparent that the modulation depth plays an important role in the $2f$ signal strength. This effect is amplified as pressure increases. For example, at 20 atm, the $2f$ signal is roughly 100% larger with $a = 0.65$ cm$^{-1}$ than with $a = 0.5$ cm$^{-1}$, whereas at 5 atm, the gain is only 30%. These results underscore the importance of large modulation depths for acquiring $2f$ signals with high SNR.

Note that for these elevated pressure conditions, the near-infrared absorption line shapes of water vapor, as shown for example in Figure 6.8, are predominantly pressure broadened and Lorentzian. Therefore, the line shapes are actually narrower at higher temperatures (for a fixed high pressure), as would be found in combustion-related high pressure gas sensing applications. The full width half maximum (FWHM), $\Delta \nu_L$, for a Lorentzian line is described by the relation:

$$
\Delta \nu_L = P \sum_j \chi_j \gamma_j^T \left( \frac{T_0}{T} \right)^{n_j} \quad (6.1)
$$
\( \Delta \nu_L \) is the Lorentzian half width at half maximum (HWHM), respectively, in wavenumber (cm\(^{-1}\)) units. \( T \) is the gas temperature in Kelvin, \( P \) is the total pressure in atmospheres, \( \chi_j \) is the mole fraction of the \( j^{th} \) component of the gas mixture, \( \gamma_j^{T_0} \) is the pressure broadening coefficient (half width) at reference temperature \( T_0 \) for the \( j^{th} \) perturbing species (cm\(^{-1}\)/atm), and \( n_j \) is the species-dependent temperature coefficient. A typical value of \( n_j \), such as for the absorption feature at 1371 nm (Figure 6.8), is 0.6. In this case, the collision width at 1000 K is half the line width at 296 K. Therefore, the 20 atm line shape simulated in Figure 6.8 and measured using 2f methods in Figure 6.7 would be similar to a 40 atm line shape at 1000 K (ignoring changes in line strength).

The rms noise, in the 2f signal units shown in Figure 6.7, is 5(10\(^{-5}\)). The 2f signal for the 20 atm water sample (0.1% mole fraction), at \( a = 0.65 \) cm\(^{-1}\), is about 5(10\(^{-3}\)). Therefore the minimum detectable water concentration at 20 atm for this experiment is approximately 0.001%, for a meter-long path length and a 15 kHz measurement bandwidth.

### 6.2.2 2f versus Temperature and Ratio Thermometry

A heated cell with a double-pass effective path length of 40 cm (Section 3.4) is used to measure 2f peak heights for two water vapor transitions at various temperatures from 296 K to 800 K at a pressure of 10 atm. The two water features probed are at 1392 nm (7185.6 cm\(^{-1}\)) and 1371 nm (7294.2 cm\(^{-1}\)). The lower-state energies of these transitions are at 1045 cm\(^{-1}\) and 23.8 cm\(^{-1}\), respectively. For temperatures in the 300-800 K range, these lines behave quite differently with respect to temperature, making them ideal candidates for ratio thermometry at these temperature conditions. Details on ratio thermometry and the relationship between lower-state energies and temperature sensitivity may be found in the literature [Arroyo and Hanson, 1993]. Comparison of the data with 2f simulations, as shown in Figure 6.9, exhibits generally good agreement. Discrepancies in the data may be attributed to errors in HITRAN, in which the broadening parameters are especially suspect. In addition, the water vapor mixtures used in these experiments were dilute, since the vapor pressure of water, about 17 Torr, was mixed with over 10 atm (7600 Torr) of nitrogen, resulting in water vapor mole fractions...
typically around 0.002. Therefore, some experimental uncertainty results from the weak absorption levels being detected, which were on the order of a few percent or less in this heated cell.

Figure 6.9: $2f$ signal versus temperature (experimental and simulated). $P = 10$ atm; $a = 0.5$ cm$^{-1}$; 1392 and 1371 nm features. Simulations utilize HITRAN2004.

Ratio thermometry is a convenient way to measure gas temperatures when gas concentration is variable or when laser transmission is unpredictable, due to beam steering or fouling effects. By taking the ratio of $2f$ signal strengths from multiplexed lasers, common-mode noise is reduced and the need to know gas concentration is eliminated (assuming a dilute sample with a relatively constant bath-gas composition). Figure 6.10 is a simulation of how the ratios of the $2f$ peak heights of the water features probed in Figure 6.9 vary with temperature. From the slope of the curve in Figure 6.10, the sensitivity of the peak ratios to changes in temperature may be calculated. For example, at 500 K, a 1% change in the peak ratios corresponds to a temperature change of about 2.5 K.
A single-point calibration at one known measurement condition is sufficient to scale the measured 2$\ell$ ratios with the simulations for the purposes of thermometry. It is necessary to have an independent measurement of total gas pressure, since the temperature sensitivity of the 2$\ell$ peak ratios is also a function of pressure. By simulating the 2$\ell$ peak heights, and thus the ratios, for two or more water features, over a matrix of relevant pressure and temperature conditions, a single calibration allows for temperature measurements even under variable pressure conditions, provided that a pressure measurement is always available. The results of this study indicate that 2$\ell$ simulations are capable of predicting and interpreting experimental results, but that more detailed spectroscopic studies are necessary in order to insure completely accurate large-modulation-depth 2$\ell$ simulations. In addition to sensitivity to line strength and lower state energy, these 2$\ell$ simulations are sensitive to line shape parameters, i.e. broadening widths and temperature coefficients, which have not been experimentally verified with great care in the literature.
6.2.3 Species Concentration

Section 2.2.3 introduces the concept behind extracting species concentration information from 2f measurements. A demonstration experiment of 2f concentration measurements of broadened spectra was conducted in the 40 cm path length heated cell (Section 3.4). The water feature at 1371 nm (7294.2 cm\(^{-1}\)) was used for this study, as it was conducted at room temperature. A dilute mixture of water vapor was introduced into the cell at room temperature and atmospheric pressure. 2f peak signals were recorded as the mixture was pressurized by the addition of compressed nitrogen. The addition of nitrogen also acts to dilute the mixture, so that the water mole fraction at each pressure point is less than the one before (\(\chi_{\text{water}} \propto 1/P\)), but is readily calculated based upon an initial measurement of water mole fraction in the original mixture. Note that the water vapor spectra may be assumed to be entirely air-broadened for all measurements due to the fact that only dilute water samples are used (less than 0.2% water). Therefore, any residual inhomogeneity in the mixture does not affect measurement accuracy, as the path integrated absorbance is equivalent to a homogeneous air-broadened mixture of the same average mole fraction. In order to infer water vapor mole fraction from the 2f signal strengths, 2f simulations are generated for the exact experimental conditions encountered, except that the simulations are based on a constant water vapor mole fraction of 0.2%. A one-point calibration is used to scale the simulations to the 2f data at the initial data point at \(P = 1.5\) atm. Clearly, as the mixture is diluted, the actual 2f signal strengths recorded become less than the simulations that are based upon a constant mole-fraction assumption. The water mole fraction is simply inferred by assuming that the 2f signal deficit, compared to simulation, is directly proportional to the reduction in water mole fraction.

Figure 6.11 shows the measured and actual water vapor mole fraction as a function of total pressure. The measured water vapor mole fraction, \(\chi^2_{\text{water}}\), is inferred from the experimental 2f signal strengths. The actual water vapor mole fraction, \(\chi_{\text{water}}\), is calculated by assuming a straightforward dilution of the initial water/nitrogen mixture (\(\chi_{\text{water}} \propto 1/P\)). Figure 6.12 is a plot of \(\chi^2_{\text{water}} / \chi_{\text{water}}\) as a function of pressure. The
maximum error in Figure 6.12 is about 3%, for a pressure range that spans nearly an order of magnitude.

Figure 6.11: Measured and actual water vapor mole fraction, $\chi_{water}$ vs. total pressure.

Figure 6.12: $\chi_{water}^{2f} / \chi_{water}$ vs. total pressure. $T = 296$ K; $a = 0.5$ cm$^{-1}$; 1371 nm (7294.2 cm$^{-1}$) water feature.
Simulations of the 1371 nm water feature used in the concentration measurements may be used to calculate a detection limit for this feature at room temperature. Here, the detection limit is defined as the mole fraction of water vapor for which a SNR of 1 exists, for a path length of 1 cm. Alternatively, as shown in Figure 6.13, we can express the detection limit in terms of the mole fraction, path length product: $\chi L$. The noise floor is determined experimentally by measuring the standard deviation in the $2f$ data using a 15 kHz lock-in filter bandwidth, a modulation frequency of 50 kHz, and a sampling rate of 5 MHz (see Section 3.3.3). In the $2f$ units shown in the plots within this work, the noise floor is $5(10)^{-5}$, as shown in Figure 3.10. Figure 6.13 indicates that the optimal minimum detectable $\chi L$ product is about $6(10)^{-5}$ for this measurement condition. In order to provide a comparison with direct absorption methods, spectral absorption line shape simulations are performed at the same conditions (1371 nm line at room temperature) for $\chi L = 6(10)^{-5}$. The results indicate that the fractional absorbance of this feature, at pressures from 1 to 10 atm, is approximately $7(10)^{-5}$. This is about an order of magnitude better than the direct absorption detection limit at 15 kHz for those conditions. Furthermore, direct absorption measurements of broadened spectra (above atmospheric pressure for water vapor) in environments with fluctuating transmission characteristics are especially challenging due to difficulties with identifying laser intensity baselines, $\bar{I}_0$. Therefore, the effective $2f$ detection limit is at least an order of magnitude improved over the comparable direct absorption detection limit.

From Figure 6.13 it is apparent that at high pressures, the modulation depth, $a$, has a strong effect on the $2f$ signal strength, and hence, the detection limit. As expected, larger modulation depths push the minimum in the detection limit curve to higher pressures. It is important to remember that $2f$ signal strengths and detection limits are dependent upon various factors, such as temperature, the specific water feature probed, pressure, and modulation depth. Line strengths, on average, generally weaken with temperature due to increasing partition functions as more energy levels are populated. Therefore, sensitive detection at elevated temperatures becomes increasingly difficult with small path lengths, though this may be mitigated through selection of absorption transitions with increased lower state energy.
Figure 6.13: Detection limit (mole fraction, path length product: $\chi L$) vs. total pressure. $T = 296$ K; 15 kHz lock-in bandwidth; 1371 nm (7294.2 cm$^{-1}$) water feature.
Chapter 7. Summary and Future Work

7.1 Summary

Tunable diode lasers have gained popularity as a technology that enables in situ, line-of-sight, optical absorption diagnostics for a wide range of applications. Whether for industrial gas sensing in power plants and plasma reactors, or environmental gas sensing, or for combustion diagnostics in engines or flames, diode-laser sensors provide non-intrusive measurements with excellent temporal resolution.

In this work, measurement campaigns in three large engine test facilities have been described, using tunable diode lasers to obtain temperature and species information by probing water vapor features in the near-infrared spectral region (1.31 – 1.47 µm). The results demonstrate the feasibility of transporting, installing, and using diode-laser sensors on large-scale engine test stands. Preliminary results show good agreement with expected combustor conditions.

First visits to a SCRAMJET facility and a high-pressure gas turbine sector rig at Wright-Patterson Air Force Base yielded insights into optical design improvements for mitigating the effects of turbulent flow field-induced beam steering noise. Refinements have also been made in the fiber-optic transmission and collection of laser radiation in order to eliminate fiber mode noise distortions. Measurements on a high-repetition-rate multi-tube pulsed detonation engine (PDE) also demonstrate the capability of the diode-laser temperature sensor to capture process timing information, such as fuel arrival, as
well as to diagnose engine mis-fires. Preliminary temperature results agree well with known vitiated fuel/air conditions and expected post-detonation temperatures.

Engine measurements elucidate and motivate a need for improved spectroscopic techniques in order to improve signal-to-noise ratios (SNR) as well as to reduce errors due to baseline laser intensity determinations that are often performed with a polynomial fit to the non-absorbing wings of isolated spectral features. Wavelength modulation spectroscopy (WMS), with second-harmonic detection ($2f$) is explored as a means to improve upon direct absorption ratio thermometry. In particular, it has been shown that the interpretation of $2f$ absorption peak ratios from multiplexed diode lasers is straightforward, provided that modulation indices are optimized, and permits accurate and fast temperature sensing over large temperature ranges with minimal calibration. For high-pressure measurements with water vapor, collisions cause spectral features to broaden and blend, removing the non-absorbing wings used to perform “baseline fits” at lower pressures. The derivative-nature of wavelength modulation spectroscopy offers major benefits in dealing with congested spectra, as the signal is sensitive to line-shape curvature rather than simply absorption magnitude.

Laboratory validation experiments in a heated optical cell have been performed using digital waveform generation and lock-in detection. Results demonstrate the accuracy of optimized $2f$ ratio thermometry when probing isolated spectra, where $2f$ peak height ratios vary directly with line-strength ratios. High-pressure measurements as a function of temperature (296 – 1000 K) and pressure (up to 20 atm) were made, utilizing large laser modulation depths. These show good agreement with $2f$ simulations based upon spectroscopic parameters tabulated in HITRAN2004.

The measurement strategies introduced here are likely to find growing use in many future measurement campaigns. $2f$ ratio thermometry shows great promise for in-cylinder temperature diagnostics in automotive engines as well as for measurements in aircraft propulsion devices. Both scanned and fixed-wavelength $2f$ strategies have been explored in this work, each offering benefits for a wide range of applications, both at low and high-pressures. Some of the implications of this research will be discussed in Section 7.2.
In conclusion, this thesis discusses various diode-laser absorption diagnostic strategies, all of which offer benefits for various measurement applications. Direct absorption methods are ideal techniques for environments with strong absorption features that are relatively isolated and narrow. For weakly-absorbing features, wavelength modulation provides an improvement to SNR as well as alleviating some uncertainties with baseline fits. The derivative nature of WMS offers benefits for spectroscopic sensing with broad and blended spectra that lack non-absorbing regions for laser intensity-baseline determinations. Fixed-wavelength methods offer fast time response but are difficult to calibrate, making them practical for diagnostics in periodic combustion processes only.

7.2 Future Work

7.2.1 SCRAMJET diagnostics

The SCRAMJET engine test facility at Wright-Patterson Air Force Base in Ohio is a prime candidate for using scanned-2f ratio thermometry with optimized modulation indices. The SCAMJET combustor has the benefit of convenient optical access through a large 1” by 6” window, and a wedged version of this window has been installed to eliminate etalons. It also runs at near-atmospheric pressures, where water vapor absorption lines are isolated and ideal for scanned-2f methods. The engine is vitiated, meaning that a preliminary combustion step is used to preheat the air flow before the main supersonic combustion process occurs. Since there is more than one combustion event, a large amount of water vapor is present in the post-combustion products (typically over 10% water fraction), ensuring ample absorption signals. Temperatures are relatively low as well (1000~2000K), which allows for good temperature sensitivity. Temperature measurements in the SCRAMJET facility aim to diagnose temporal fluctuations through fast (kilohertz-rate) sensing as well as to monitor spatial pattern factors. An existing translation stage on both sides of the SCRAMJET optical access window provides the
ability to translate the sensor beam path along one cross-section of the engine for the purpose of measuring spatial temperature profiles.

The challenge for implementing $2f$ ratio thermometry in a SCRAMJET engine is calibrating the sensor. Unlike periodic combustion devices such as an internal combustion engine or a pulsed detonation engine, there is not a constant reference, such as the inlet fuel/air condition, for the sensor to be calibrated constantly. In a continuously-operating engine, signals may drift due to mechanically or thermally-induced misalignments. One solution is to combine direct-absorption with WMS. Direct absorption may be intermittently carried out to provide an absolute and calibration-free measurement of temperature and species mole fraction. WMS would then provide fast measurements of relative temperature or concentration fluctuations at all other times. For certain applications, monitoring relative changes in temperature or concentration, over time or space, may be of more value than knowing absolute values. In these cases, accurate calibration is much less important.

7.2.2 High-Pressure Combustion Diagnostics

Development of the large-modulation-depth $2f$ thermometry strategy was motivated by a desire to rapidly monitor gas temperatures at high pressures. Absorption levels in realistic combustion devices are often weak, due to limited path length or absorbing species concentration, so that modulation spectroscopy is necessary for the increased SNR it affords. A study of optimal spectroscopic line choices has been made for a particular measurement goal, following a HITRAN-based selection procedure similar to the one described in Zhou, et al. (2003). Future work includes the confirmation and improvement of spectroscopic parameters for the chosen line selections, by detailed laboratory measurements in a high-pressure optical cell heated with a high-uniformity three-zone tube furnace (Micropyretics Heaters International). Trustworthy spectroscopic parameters are necessary to perform accurate $2f$ spectral simulations, over the matrix of temperatures and pressures relevant for a specific high-pressure measurement application. $2f$ simulations must then be compared with high-pressure $2f$
measurements in a heated cell, in order to verify that simulations are accurate and suitable for interpreting 2f signal strengths for thermometry and species.

One concern that must be addressed is the effect of pressure-shift on the interpretation of 2f data. Pressure-shift coefficients are not accurately known, and must be carefully determined experimentally. Simulations indicate that pressure-shift has a significant effect on 2f signal strengths and 2f peak height ratios, over the wide range of pressures common in high-pressure combustion devices. Simulations also indicate that the effect of variations in gas composition has a non-trivial effect on 2f signals and ratios. This is due to the fact that the collisional broadening coefficient due to air is quite different from the collisional broadening due to self-colliders (water vapor). Therefore, as water vapor concentrations vary, due to fluctuations in absorber concentration, slight adjustments must be made to interpret 2f signal ratios and magnitudes for thermometry and species concentration measurements.

A fully-assembled sensor system requires testing and design iterations in order to eliminate an assortment of noise sources. Testing may be performed in a shock-tube facility, which provides well-known pressure and temperature conditions for validating sensor accuracy. Final testing in a realistic device would provide a valuable demonstration of this sensor strategy reduced to practice.

7.2.3 Investigating Higher Harmonics for WMS

While second-harmonic detection is by far the most popular form of demodulation used in practical WMS systems, some have suggested that there are benefits to employing higher-harmonic detection. For example, Dharamsi et al. (1998) show that etalon effects from parallel optical surfaces in the diagnostic apparatus are reduced when analyzing higher harmonics. Dharamsi and Bullock also show that detection at higher harmonics, or a combination of higher harmonics, allows for improved resolution and detection of broadened spectral features (Dharamsi and Bullock, 1996; Bullock and Dharamsi, 1998). Their work did not focus on practical sensing applications, but some analytical and/or simulation work could be executed to
demonstrate valuable benefits and improvements to overall SNR for some measurement applications.

7.2.4 Other Measurement Applications

2f ratio thermometry and wavelength modulation techniques in general, have broad applicability for many measurement applications. Attempts are being made to incorporate WMS in pulsed detonation engine diagnostics, where calibrations may be performed on a cycle-by-cycle basis based upon known inlet fuel/air fill conditions. Another potential high-pressure measurement test bed is a gas-turbine combustor. As with the SCRAMJET application discussed in Section 7.2.1, more thought must go into determining how to calibrate a sensor utilizing WMS if the combustion process is continuous as with a gas turbine. Additional measurement applications are endless: environmental monitoring, biological and biomedical diagnostics and imaging, power plant diagnostics, sensors for semiconductor processing, etc. While this work was focused on water vapor measurements of temperature and concentration, there are other species and other parameters of interest that may be interrogated using similar methods, such as flow velocity using Doppler-shift and time-of-flight techniques. Finally, the unique extension of WMS to blended and broadened spectra, using large modulation depths, opens up the possibility of measuring larger molecules such as hydrocarbon fuels, which possess congested spectra even at low pressures.
Appendix A. Additional Data

The following data were taken to verify candidate water vapor features to determine an optimal line selection for an in-cylinder sensor to monitor temperature during the compression stroke of an internal combustion engine. Candidate lines were selected from direct-absorption and $2f'$ spectral simulations, for expected compression stroke conditions, based on the HITRAN2004 database. The selection process was similar to the one described by Zhou, et al. (2003).

Note that nitrogen is used as the bath gas in experimental studies whereas all simulations are based upon air-broadening coefficients. Therefore, since nitrogen is about 10% more efficient than air as a broadening species for water vapor in the NIR (approximately), one would expect the experimental data to be slightly broadened compared to the simulations in Section A.1. Because these validations were designed as rough comparisons between simulation and experiment, the plots are left uncorrected for this effect.

Section A.1 contains plots of direct absorption scans acquired at specific temperature and pressure conditions in a heated optical cell (Section 3.4). Comparison of these absorbance plots with simulated spectra allows for a quick check that HITRAN2004 parameters, such as line strength and broadening coefficients, are reasonably accurate. Section A.2 contains plots of fixed-wavelength $2f'$ line-center signals at a single temperature condition, as a function of pressure (up to about 10 atm). These data points are normalized at 1 atm in order to scale them with the simulations.
Measuring 2f peak heights as a function pressure allows for a quick check of HITRAN2004 broadening coefficients.

Table A.1 lists the candidate water vapor features that were investigated as potential line choices for a high pressure temperature sensor. The majority of the measurements indicate good agreement with HITRAN2004 simulations. However, a few lines exhibit significant discrepancies. In the case of the feature at 1344.9 nm ($E'' = 1558$ cm$^{-1}$), the data is more favorable than the simulations. The feature at 1434.15 nm ($E'' = 1558$ cm$^{-1}$) broadens more with pressure than the simulations predict, and is not a good choice for high-pressure diagnostics.

Table A.1: Summary of water vapor features investigated for the development of a high-pressure temperature sensor

<table>
<thead>
<tr>
<th>Absorption peak wavelength; nm (cm$^{-1}$)</th>
<th>Lower state Energy, $E''$; cm$^{-1}$</th>
<th>Agreement with simulation (HITRAN2004)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1343.3 (7444.4)</td>
<td>1807</td>
<td>Similar</td>
</tr>
<tr>
<td>1344.9 (7435.6)</td>
<td>1558</td>
<td>Not similar (better)</td>
</tr>
<tr>
<td>1346.6 (7426.2)</td>
<td>1327</td>
<td>Similar</td>
</tr>
<tr>
<td>1350.4 (7405.1)</td>
<td>920</td>
<td>Similar</td>
</tr>
<tr>
<td>1352.5 (7393.8)</td>
<td>744</td>
<td>Similar</td>
</tr>
<tr>
<td>1385.1 (7219.6)</td>
<td>488</td>
<td>Similar</td>
</tr>
<tr>
<td>1388.4 (7203.9)</td>
<td>742</td>
<td>Similar</td>
</tr>
<tr>
<td>1395.7 (7164.9)</td>
<td>1395</td>
<td>Similar</td>
</tr>
<tr>
<td>1434.15 (7972.8)</td>
<td>1558</td>
<td>Not similar (worse)</td>
</tr>
</tbody>
</table>
A.1 Direct Absorption Comparisons with Simulation

Figure A.1: Absorbance versus frequency (wavenumber units). Water vapor in nitrogen (~0.15%). $\lambda_{peak} = 1343.3$ nm (7444.4 cm$^{-1}$), $E'' = 1807$ cm$^{-1}$, $T = 762$ K, $L = 40.5$ cm, $P = 5671$ Torr

Figure A.2: Absorbance versus frequency (wavenumber units). Water vapor in nitrogen (~0.15%). $\lambda_{peak} = 1344.9$ nm (7435.6 cm$^{-1}$), $E'' = 1558$ cm$^{-1}$, $T = 595$ K, $L = 40.5$ cm, $P = 5555$ Torr
Figure A.3: Absorbance versus frequency (wavenumber units). Water vapor in nitrogen (~0.15%). $\lambda_{\text{peak}} = 1346.6$ nm ($7426.2$ cm$^{-1}$), $E'' = 1327$ cm$^{-1}$, $T = 595$ K, $L = 40.5$ cm, $P = 5555$ Torr.

Figure A.4: Absorbance versus frequency (wavenumber units). Water vapor in nitrogen (~0.15%). $\lambda_{\text{peak}} = 1350.4$ nm ($7405.1$ cm$^{-1}$), $E'' = 920$ cm$^{-1}$, $T = 595$ K, $L = 40.5$ cm, $P = 5555$ Torr.
Figure A.5: Absorbance versus frequency (wavenumber units). Water vapor in nitrogen (~0.15%). $\lambda_{peak} = 1352.5$ nm (7393.8 cm$^{-1}$), $E'' = 744$ cm$^{-1}$, $T = 296$ K, $L = 40.5$ cm, $P = 5980$ Torr.

Figure A.6: Absorbance versus frequency (wavenumber units). Water vapor in nitrogen (~0.15%). $\lambda_{peak} = 1385.1$ nm (7219.6 cm$^{-1}$), $E'' = 488$ cm$^{-1}$, $T = 296$ K, $L = 40.5$ cm, $P = 6372$ Torr.
Figure A.7: Absorbance versus frequency (wavenumber units). Water vapor in nitrogen (~0.15%). $\lambda_{\text{peak}} = 1395.7$ nm (7164.9 cm$^{-1}$), $E'' = 1395$ cm$^{-1}$, $T = 693$ K, $L = 40.5$ cm, $P = 3800$ Torr

Figure A.8: Absorbance versus frequency (wavenumber units). Water vapor in nitrogen (~0.15%). $\lambda_{\text{peak}} = 1434.15$ nm (7972.8 cm$^{-1}$), $E'' = 1558$ cm$^{-1}$, $T = 296$ K, $L = 40.5$ cm, $P = 7490$ Torr
A.2 2f vs Pressure Compared with Simulations

Figure A.9: Normalized (point calibration) 2f peak height signal versus total pressure. Water vapor in nitrogen (~0.15%). $\lambda = 1344.9$ nm (7435.6 cm$^{-1}$), $E'' = 1558$ cm$^{-1}$, $T = 608$ K, $a = 0.6$ cm$^{-1}$

Figure A.10: Normalized (point calibration) 2f peak height signal versus total pressure. Water vapor in nitrogen (~0.15%). $\lambda = 1346.6$ nm (7426.2 cm$^{-1}$), $E'' = 1327$ cm$^{-1}$, $T = 608$ K, $a = 0.6$ cm$^{-1}$
Figure A11: Normalized (point calibration) $2f$ peak height signal versus total pressure. Water vapor in nitrogen (~0.15%). $\lambda = 1350.4 \text{ nm} (7405.1 \text{ cm}^{-1})$, $E'' = 920 \text{ cm}^{-1}$, $T = 608 \text{ K}$, $a = 0.6 \text{ cm}^{-1}$

Figure A.12: Normalized (point calibration) $2f$ peak height signal versus total pressure. Water vapor in nitrogen (~0.15%). $\lambda = 1385.1 \text{ nm} (7219.6 \text{ cm}^{-1})$, $E'' = 488 \text{ cm}^{-1}$, $T = 296 \text{ K}$, $a = 0.6 \text{ cm}^{-1}$
Figure A.13: Normalized (point calibration) $2f$ peak height signal versus total pressure. Water vapor in nitrogen (~0.15%). $\lambda = 1388.4$ nm (7203.9 cm$^{-1}$), $E'' = 742$ cm$^{-1}$, $T = 488$ K, $a = 0.6$ cm$^{-1}$

Figure A.14: Normalized (point calibration) $2f$ peak height signal versus total pressure. Water vapor in nitrogen (~0.15%). $\lambda = 1395.7$ nm (7164.9 cm$^{-1}$), $E'' = 1395$ cm$^{-1}$, $T = 773$ K, $a = 0.6$ cm$^{-1}$
Figure A.15: Normalized (point calibration) $2f$ peak height signal versus total pressure. Water vapor in nitrogen (~0.15%). $\lambda = 1434.2$ nm ($6972.8$ cm$^{-1}$), $E'' = 1558$ cm$^{-1}$, $T = 773$ K, $a = 0.6$ cm$^{-1}$
References


http://cfa-www.harvard.edu/hitran/


