Development and Application of High-Speed Raman/Rayleigh Scattering in Turbulent Nonpremixed Flames

DISSERTATION

Presented in Partial Fulfillment of the Requirements for the Degree Doctor of Philosophy in the Graduate School of The Ohio State University

By

Kathryn N. Gabet Hoffmeister

Graduate Program in Mechanical Engineering

The Ohio State University

2015

Dissertation Committee:

Dr. Jeffrey A. Sutton, Advisor
Dr. Walter R. Lempert
Dr. Mohammad Samimy
Dr. Shaurya Prakash
Abstract

In this dissertation, a new, high-speed, combined 1D Raman-Rayleigh scattering imaging approach was developed for quantitative temporally-correlated (10-kHz) measurements of temperature, major combustion species (O\textsubscript{2}, N\textsubscript{2}, H\textsubscript{2}O, and H\textsubscript{2}), and mixture fraction in the turbulent DLR H3 nonpremixed jet flame. The new high-speed measurements presented here were facilitated through the development of a custom high-speed imaging spectrometer, implementation of a robust data reduction methodology, and the use of the High-Energy Pulse Burst Laser System (HEPBLS) at Ohio State, which produces ultra-high pulse energies at multi-kHz repetition rates. Detailed measurements in near-adiabatic, laminar calibration flames were used to assess the accuracy and precision of the kHz-rate measurements using the high-speed Raman/Rayleigh scattering imaging system. In general, good agreement was found as compared to adiabatic flame calculations over a broad range of temperature and equivalence ratios.

Current 10-kHz measurements and derived statistics within the turbulent H3 flames were compared to previously-measured, low-repetition-rate scalar data available through the Turbulent Nonpremixed Flame (TNF) workshop database. In general, good agreement between the mean values and RMS fluctuations of the temperature and major species were found between the current and previous data, indicating sufficient accuracy in single-shot measurements in turbulent environments. Following technique
development, a series of 10-kHz measurements were used to visualize the highly intermittent dynamics of the scalar fields. In addition to flow visualization, time-dependent measurements were used to deduce the temporal autocorrelation function and the associated integral time-scales of the major species, mixture fraction, and temperature in the DLR H3 flames. This research presents the first measurements of the integral timescales of the major combustion species and mixture fraction in turbulent flames as well as the first reporting of integral timescales of multiple scalars simultaneously. While the integral timescales of all scalars (O₂, H₂O, and H₂, ξ, and T) generally increase with both axial and radial position, the individual integral timescales are significantly different, displaying a factor of three spread across all of the measured scalars. The integral timescales for temperature and water are highly correlated to one another at all spatial locations, while the integral timescale for mixture fraction closely tracks that of hydrogen between the jet centerline and the stoichiometric contour and tracks that of oxygen between the stoichiometric contour and the co-flowing oxidizer stream. Results indicate that in regions of high chemical activity and heat release, the use of single characteristic (integral) timescale to describe the large-scale behavior is not appropriate; that is, each scalar has its own unique spatially-dependent integral timescale. For spatial positions beyond the flame tip, the integral timescales for all scalars collapse upon one another meaning that the system can be described adequately as a pure mixing situation with a single characteristic timescale. Finally, new, temporal cross-correlations of various scalar pairs are presented along with a discussion of the derived scalar interaction times. In general, the scalar interaction times are bound by the individual scalar integral
timescales, although unique characteristics are observed near the stoichiometric contour, which varies amongst the various scalar pairs. It is expected that the newly developed high-speed 1D Raman/Rayleigh imaging approach will provide new physical insight into the intermittent behavior of turbulent nonpremixed combustion and the subsequent turbulence-chemistry interaction, as well as providing new, time-resolved data for assessment and validation of time-dependent combustion models.
To my family for their hugs, words of encouragement, and dirty looks when necessary.
Acknowledgments

I would like to thank my family for their support throughout this very long journey. Mom and Dad – thank you for raising me to believe in myself and being there to reinforce the message when necessary. Heather, Kurt, Josh, and Mariel – thank you for your encouragement and understanding. Thank you also for the photos of your adorable children to make me smile when I needed it most. My in-laws, aunts, uncles, and cousins – thank you so much for always asking when I am graduating as though it was a sure-thing.

Brian – thank you for your patience, your understanding, and your help along the way. I likely wouldn’t have finished this degree without you by my side to buy me ice cream to celebrate my victories, to draw cartoons of me triumphing when things weren’t going my way, and to make sure I ate more than vending machine snacks when I was working late at night. Thank you for all the things you have done for me, large and small.

I would like to thank my friends and coworkers for all the lunches, dinners, workouts, Facebook posts, and weekends of fun to take my mind off of work. You made my days brighter and the long nights easier. Emily Sequin, Karen Bellman, Aarthy Vaidyanathan, Marie Fuest, Sherrie Bowman, Suzanne Sheehe, Krista Kecskemety, Michelle Lockhart, and Erin Hutter – thank you for being there to get coffee, listening, and making sure that I knew I wasn’t alone. Randy Patton and Ting Li – thank you for
your help and comraderie. I couldn’t have asked for better people to help me figure things out. Whether it was homework or how to run fuel lines, I could always count on you guys to trudge through with me. Sherrie Bowman, Yvette Zuzeek, Matt Webster, and Naibo Jiang – thank you for explaining to me how lasers work, how not to burn myself with one, where to buy the things I need to keep from burning myself, etc. Frederik and Mike – thank you for helping me take data so that I could finally graduate. The rest of you – thank you for all the jokes, baked goods, borrowed equipment, and other things that helped along the way.

Thank you also to my collaborators – Rob Barlow, Matt Dunn, and Frederik Fuest for patiently answering my many, many questions about Raman data processing and to Dirk Geyer and Andreas Dreizler for providing the RAMSES code.

Thank you to the DOE SCGF for my fellowship, specifically Ping Ge and Cayla Stephenson for all your hard work.

I would also like to thank the staff in the Mechanical and Aerospace Engineering Department at Ohio State for their help over the years. Nick Breckenridge and Janeen Sands – thank you for your advice, encouragement, and willingness to work out all the bureaucratic problems I encountered. Dr. Lempert, Dr. Samimy, Dr. Prakash, Dr. Siston, Dr. Mazumder, Dr. Heremans, and others – thank you for everything you have taught me about engineering, teaching, and surviving graduate school.

Dr. Sutton – thank you for your guidance and patient explanations. This has been a very long journey and you have taught me so much. Thank you for believing in me.
Vita

May 2004 ..................................................Jackson High School, Massillon, OH

May 2008 ..................................................B.S. Aerospace Engineering, Case Western Reserve University

Feb 2012 ..................................................M.S. Mechanical Engineering, The Ohio State University

2012 to present ........................................Graduate Fellow, Department of Mechanical and Aerospace Engineering, The Ohio State University

Publications


Fields of Study

Major Field: Mechanical Engineering

Areas of Interest: Laser Diagnostics, Combustion, Turbulence, Energy Efficiency
Table of Contents

Abstract ............................................................................................................................................. ii

Acknowledgments ........................................................................................................................ vi

Vita ................................................................................................................................................ viii

Table of Contents .......................................................................................................................... x

List of Tables .................................................................................................................................. xiv

List of Figures ................................................................................................................................. xvi

Chapter 1: Introduction and Overview ......................................................................................... 1

1.1 Motivation ................................................................................................................................. 1

1.2 Scope of Research .................................................................................................................... 4

1.3 Development of High-Speed 1D Raman/Rayleigh Imaging .................................................. 6

1.4 Characterization of Turbulence-Chemistry Interaction ............................................................ 7

1.5 Dissertation Outline ................................................................................................................ 9

Chapter 2: Background .................................................................................................................. 11

2.1 Mixture Fraction ..................................................................................................................... 11

2.2 Relevant Length and Time Scales ........................................................................................... 13
2.3 Laser Diagnostics ................................................................. 15
2.4 Raman and Rayleigh Scattering .............................................. 17
2.5 Applications of Raman and Rayleigh Scattering in Combustion Environments .... 21
2.6 High Speed Imaging.............................................................. 24

Chapter 3: Experimental Methods .............................................. 28
3.1 Signal Collection .................................................................. 30
3.2 Laser Excitation Systems ...................................................... 34
3.2.1 10-Hz Q-Switched Nd:YAG Laser ........................................... 34
3.2.2 High-Energy Pulse Burst Laser System ................................. 35
3.3 Pulse Stretcher .................................................................. 41
3.4 Burners and Test Facilities ..................................................... 45
3.4.1 McKenna Flat-Flame Burner ................................................... 45
3.4.2 Near-Adiabatic Hencken Burner ................................................. 46
3.4.3 Turbulent Jet Flame in Co-Flow ............................................... 48
3.5 Cameras and Detection System ............................................... 51
3.6 Spatial Resolution and Sampling Rates ...................................... 60

Chapter 4: Data Processing .......................................................... 65
4.1 Preprocessing .................................................................... 66
4.1.1 Camera Mapping ............................................................... 66
7.3 Scalar Cross-correlations and Timescales......................................................... 197

Chapter 8: Summary, Conclusions, and Suggested Future Work.............................. 211

References............................................................................................................. 218
List of Tables

Table 2.1: Species scattering cross-sections at 295 K ......................................................... 21
Table 3.1: Coefficients of determination ($R^2$) of linear fit to measured camera signal response......................................................................................................................... 55
Table 3.2: Calculated Batchelor length scales and frequencies in the turbulent 50% N$_2$/50% H$_2$ jet flame issuing into air at Re=10,000 .......................................................... 64
Table 4.1: Wavelengths and corresponding pixel numbers used to map the Raman-camera pixels to spectral positions........................................................................................................ 71
Table 4.2: Spectral binning used on the Raman camera to match conditions at Sandia National Laboratories....................................................................................................................... 72
Table 4.3: Example Raman-channel signal verification for a non-reacting flow of 0.59 N$_2$ / 0.16 O$_2$ / 0.25H$_2$. ...................................................................................................................... 85
Table 4.4: Raman channel progression ..................................................................................... 93
Table 4.5: RAMSES active matrix locations ........................................................................... 97
Table 5.1: CEA calculations for near-adiabatic calibration flames ........................................ 99
Table 5.2: Representative signal-to-noise ratios in near-adiabatic calibration flames from measurements taken at the CRF at Sandia National Laboratories .................... 112
Table 5.3: Representative signal-to-noise ratios in near-adiabatic calibration flames at 10 Hz ................................................................................................................................. 117
Table 5.4: Representative signal-to-noise ratios in near-adiabatic calibration flames at 10 kHz.
List of Figures

Figure 2.1: Energy levels diagram for Rayleigh and Raman transitions .................. 18

Figure 3.1: Combined 1D Raman/Rayleigh scattering imaging setup in the Turbulence and Combustion Research Laboratory at Ohio State University ......................... 29

Figure 3.2: Photographs of the 1D Raman/Rayleigh scattering setup ....................... 30

Figure 3.3: Raman/Rayleigh experimental setup .................................................. 32

Figure 3.4: Schematic of the High-Energy Pulse Burst Laser System (HEPBLS) .......... 37

Figure 3.5: Photographs of High-Energy Pulse Burst Laser System (HEPBLS) .......... 38

Figure 3.6: Normalized spatial profile of HEPBLS output beam as a function of pulse number within a single burst ................................................................. 39

Figure 3.7: Spatial profile of HEPBLS ................................................................. 40

Figure 3.8: Output energy as a function of burst pulse number in two sample HEPBLS pulse bursts ................................................................................................. 40

Figure 3.9: A schematic diagram of the three-leg pulse stretcher .............................. 42

Figure 3.10: The temporal profile of the output from a 15-ns (1/e^2 value) 10-Hz pulse .. 42

Figure 3.11: Rayleigh image and spatial profile of the 10-kHz beam after the implementation of a 1-leg pulse stretcher ......................................................... 44

Figure 3.12: Photograph of McKenna burner and energy measurement camera ........ 45

Figure 3.13: Hencken Burner schematic from [120] ................................................. 47
Figure 3.14: Image of a H$_2$-air Hencken flame at $\phi=0.7$ ................................................. 47
Figure 3.15: Turbulent jet in-coflow-facility ........................................................................ 49
Figure 3.16: Image of a DLR H3 (Re=10,000) jet flame with green dashes depicting the 10 kHz measurement locations at x/D=10, 20, and 40 ....................................................... 50
Figure 3.17: Photographs of the Rayleigh and Raman camera setup. ................................. 52
Figure 3.18: Red-enhanced vs. blue-enhanced photocathode quantum efficiency .......... 55
Figure 3.19: Camera linearity study ..................................................................................... 56
Figure 3.20: Intensifier charge depletion study .................................................................... 59
Figure 4.1: Spatial mask used for mapping Raman and Rayleigh cameras ....................... 67
Figure 4.2: Spatial mask images in the Raman and Rayleigh cameras ............................ 68
Figure 4.3: Typical spectral output of Newport 6032 neon lamp taken from Newport Technical Reference sheets .................................................................................................. 70
Figure 4.4: Results from a RAMSES fit to measured Raman spectra in air at 300 K using the Rayleigh signal in room-temperature air as the instrument function ....................... 76
Figure 4.5: A comparison of the measured and predicted Raman signal of nitrogen for various incident signals ........................................................................................................... 78
Figure 4.6: Results from RAMSES fit to measure Raman spectra in air at 300 K .......... 80
Figure 4.7: Signal-to-noise ratio (SNR) of nitrogen with varying spatial resolution via software binning ............................................................................................................... 83
Figure 4.8: Signal vs. HEPBLS shot number averaged over ten pulse bursts in air and corrected for laser energy ............................................................................................................ 87
Figure 4.9: Temperature vs. shot number of air in a single burst as calculated using the hybrid method .......................................................... 87
Figure 4.10: Sketch of potential signal bleeding subtraction from background effects. .. 92
Figure 4.11: 10 x 10 Raman response matrix (C) used in this research .................. 97
Figure 5.1: Matrix inversion results in air at 297 K.............................................. 101
Figure 5.2: Matrix inversion results in a non-reacting H₂/air flow at 297K.............. 102
Figure 5.3: Species concentrations and temperature in H₂/Air calibration flames as a function of equivalence ratio after a first iteration of calibrations......................... 104
Figure 5.4: Species concentrations and temperature in H₂/Air calibration flames as a function of equivalence ratio after a second iteration of calibrations....................... 106
Figure 5.5: Species concentrations and temperature in H₂/Air calibration flames as a function of equivalence ratio after a third iteration of calibrations. ......................... 107
Figure 5.6: Species concentrations and temperature in H₂/Air calibration flames as a function of equivalence ratio after a fourth iteration of calibrations ..................... 110
Figure 5.7: Measured temperature and mole fractions in near-adiabatic flat flames at the Combustion Research Facility (CRF) at Sandia National Laboratories ................. 113
Figure 5.8: Measured temperature and mole fractions in near-adiabatic flat flames at 10 Hz.................................................................................................................. 116
Figure 5.9: Measured temperature and mole fractions in near-adiabatic flat flames at 10 kHz.................................................................................................................. 119
Figure 5.10: Signal-to-noise ratios of species mole fractions in near-adiabatic, laminar flames as a function of equivalence ratio...................................................... 124
Figure 5.11: Signal-to-noise ratios of species mole fractions in near-adiabatic, laminar flames as a function of equivalence ratio................................................................. 127

Figure 5.12: Signal-to-noise ratios of species concentrations in near-adiabatic, laminar flames as a function of equivalence ratio................................................................. 128

Figure 5.13: Signal-to-noise ratio as a function of equivalence ratio for Rayleigh temperature and nitrogen mole fraction................................................................. 131

Figure 5.14: Probability density functions (PDF) of temperature and species mole fractions in non-adiabatic, laminar flames plotted versus equivalence ratio.............. 132

Figure 5.15: Estimated inaccuracies in near-adiabatic flames................................. 135

Figure 6.1: Radial profiles of the mean and RMS fluctuations of major species mole fraction and temperature in the H3 flame ......................................................... 139

Figure 6.2: Radial profile of the mean and RMS fluctuations of O₂ mole fraction at x/D=10 ..................................................................................................................... 140

Figure 6.3: Scatterplots of the major species mole fraction and temperature in mixture fraction space for the H3 flame................................................................. 143

Figure 6.4: Conditionally-averaged values of major species mole fraction and temperature in the H3 flame ................................................................. 144

Figure 6.5: Probability density functions of temperature and species mole fractions near stoichiometric conditions at x/D=20 ................................................................. 147

Figure 6.6: Probability density functions of temperature and species mole fractions for lean mixture fraction conditions at x/D=20 ................................................................. 148
Figure 6.7: Probability density functions of temperature and species mole fractions for rich mixture fraction conditions at x/D=20

Figure 6.8: Probability density functions of temperature and species mole fractions for near stoichiometric mixture fraction conditions at x/D=40

Figure 6.9: Radial profiles of the mean and RMS fluctuations of major species mole fraction and temperature in the H3 flame

Figure 6.10: Scatterplots of the major species mole fractions and temperature in mixture fraction space for the H3 flame

Figure 6.11: Conditionally-averaged values of major species mole fraction and temperature in the H3 flame

Figure 6.12: Probability density functions of temperature and species mole fractions near stoichiometric conditions at x/D=20

Figure 6.13: Probability density functions of temperature and species mole fractions for lean mixture fraction conditions at x/D=20

Figure 6.14: Probability density functions of temperature and species mole fractions for rich mixture fraction conditions at x/D=20

Figure 6.15: Probability density functions of temperature and species mole fractions for near stoichiometric mixture fraction conditions at x/D=40

Figure 7.1: Sample 10-kHz Raman scattering images acquired using the high-speed 1D Raman/Rayleigh scattering system at Ohio State

Figure 7.2: Sample 10-kHz Raman scattering images acquired using the high-speed 1D Raman/Rayleigh scattering system at Ohio State
Figure 7.3: Raman scattering images acquired at an axial position of x/D=10 and centered at a radial position of 3 mm from centerline using the high-speed 1D Raman/Rayleigh scattering system at Ohio State ................................................................. 170

Figure 7.4: Raman scattering images acquired at an axial position of x/D=20 and centered at a radial position of 4 mm from centerline using the high-speed 1D Raman/Rayleigh scattering system at Ohio State ................................................................. 171

Figure 7.5: Raman scattering images acquired at an axial position of x/D=20 and centered at a radial position of 13.3 mm from centerline using the high-speed 1D Raman/Rayleigh scattering system at Ohio State ................................................................. 172

Figure 7.6: Raman scattering images acquired at an axial position of x/D=40 and centered at a radial position of 2.5 mm from centerline using the high-speed 1D Raman/Rayleigh scattering system at Ohio State ................................................................. 173

Figure 7.7: 2D Spatio-temporal map of scalar data acquired at x/D=20 and centered at a radial position of 2.5 mm ................................................................. 176

Figure 7.8: 2D Spatio-temporal map of scalar data acquired at x/D=20 and centered at a radial position of 12 mm ................................................................. 177

Figure 7.9: Single 10-kHz time trace of temperature and species mole fraction acquired at an axial position of x/D=20 and a radial position of 1.5 mm from the centerline ...... 178

Figure 7.10: Single 10-kHz time trace of temperature and species mole fraction acquired at an axial position of x/D=20 and a radial position of 12 mm from the centerline ...... 179
Figure 7.11: 10-kHz scalar fluctuations (in mixture fraction space) within a single 15-ms burst at an axial position of x/D=20 and a radial position of r=12 mm from the centerline .................................................................................................................................................. 181

Figure 7.12: Average autocorrelation functions of temperature, major species, and mixture fraction at x/D=20 ................................................................................................................................................. 184

Figure 7.13: Average autocorrelation functions for temperature, major species, and mixture fraction at various radial positions and an axial position of x/D=20.............. 187

Figure 7.14: Calculated integral time scales (τ) at x/D=20 as a function of radial position ..................................................................................................................................................... 190

Figure 7.15: Average autocorrelation functions of temperature, major species, and mixture fraction at x/D=40 ................................................................................................................................................. 192

Figure 7.16: Average autocorrelation functions for temperature, major species, and mixture fraction at various radial positions and an axial position of at x/D=40............ 193

Figure 7.17: Average autocorrelation functions for temperature, major species, and mixture fraction at a radial position of 1 mm from the centerline ................................. 194

Figure 7.18: Average autocorrelation functions for temperature, major species, and mixture fraction at a radial position of 11 mm from the centerline......................... 195

Figure 7.19: Calculated integral time scales (τ) at x/D=40 as a function of radial position ..................................................................................................................................................... 196

Figure 7.20: Average cross-correlation functions for hydrogen mole fraction and remaining scalars at various radial positions and an axial position of x/D=20 ............ 199
Figure 7.21: Average cross-correlation functions for oxygen mole fraction and remaining scalars at various radial positions and an axial position of x/D=20................................. 200

Figure 7.22: Average cross-correlation functions for water mole fraction and remaining scalars at various radial positions and an axial position of x/D=20................................. 202

Figure 7.23: Average cross-correlation functions for temperature and remaining scalars at various radial positions and an axial position of x/D=20 ..................................................... 203

Figure 7.24: Cross-scalar time scales as a function of radial position at x/D=20........ 205

Figure 7.25: Average cross-correlation functions for oxygen mole fraction and remaining scalars at various radial positions and an axial position of x/D=40................................. 207

Figure 7.26: Average cross-correlation functions for water mole fraction and remaining scalars at various radial positions and an axial position of x/D=40................................. 208

Figure 7.27: Average cross-correlation functions for temperature and remaining scalars at various radial positions and an axial position of x/D=40 ..................................................... 209

Figure 7.28: Scalar-interaction timescales as a function of radial position at x/D=40... 210
Chapter 1: Introduction and Overview

1.1 Motivation

Despite growing concerns over the impacts of climate change, rising oil prices, and conflicts in the Middle East, over 80% of the total global energy supply is derived from the burning of fossil fuels. While this dependence on fossil fuels is expected to decline over the coming decades, estimates for 2035 continue to predict fossil fuels having a 60-80% share in energy production [1], making the development of more efficient energy-conversion systems a priority. Much of this progress will undoubtedly come from a better fundamental understanding of the turbulent combustion processes occurring within internal combustion and gas-turbine engines used in the transportation and power-production sectors [1], [2]. In addition, it is expected that the replacement of traditional prototype testing with computer-based design and optimization could lead to significant advances in performance. In this manner, a Department of Energy workshop on Basic Research Needs for Clean and Efficient Combustion of 21st Century Transportation Fuels recently recommended one primary goal for researchers [2]:

The development of a validated, predictive, multi-scale, combustion modeling capability to optimize the design and operation of evolving fuels in advanced engines for transportation applications.
Just as predictive aerodynamic models have revolutionized body design of aircraft and ground vehicles, predictive combustion models have the potential to fundamentally change how engines are developed by reducing the time and cost associated with integrating new technologies into advanced designs [2], [3]. The development of predictive models is dependent not only on improvements in computational strategies, but also progress in the understanding of the underlying physics they should describe. At moderate to high Reynolds numbers, much of the burden of describing the fundamental combustion physics and assessing modeling strategies such as large-eddy simulation (LES) falls to experimental research. Measurements of flame temperature, species concentrations, pressure, density, and velocity can be used to provide a quantitative understanding of the relationship between turbulence and flame chemistry. In addition, these measurements can be used to provide detailed databases for model assessment and validation. Gaining an understanding of the basic chemical mechanisms, the governing fluid dynamic processes, and their coupling through the use of laboratory-scale experiments in canonical configurations is an important step necessary to developing models capable of predictive design, which in turn, are aimed at improving flame efficiency, stability, and reduced emissions (particularly NO\textsubscript{x} formation) in large-scale combustion systems.

To this end, the International Workshop on Measurement and Computation of Turbulent Nonpremixed Flames (TNF Workshop) serves as one example of an approach to accelerate the development of predictive model capabilities. The TNF workshop was established to provide a biannual forum for the identification of priorities for
experimental and computational research on turbulent flames. In addition to research updates and information exchange, the TNF Workshop curates an internet library of velocity and multi-scalar data from a series of canonical turbulent flame configurations (i.e. “target flames”) to advance the basic scientific understanding of turbulent combustion and encourage collaborative comparisons of experimental data and model predictions. However, the majority of available data was measured at low acquisition rates (< 10 Hz) and provides only low-order statistics (i.e. mean, standard deviation, conditional averages) for comparisons to models. While the overarching approach of the TNF is widely regarded as successful within the turbulent combustion community, recent reports from the TNF workshop proceedings and turbulent combustion modeling review articles [4], [5] indicate a significant need to move beyond simple low-order statistical (i.e. mean and RMS) evaluation of modeling results (in particular, those arising from time-dependent LES calculations). Strong arguments have been presented for the utility of time-series measurements in assessing model performance under transient and non-stationary conditions, in addition to providing new insights into time-dependent combustion physics (discussed in greater detail in Section 2.5).

The goal of the research presented in this dissertation is to develop a temporally-resolved 1D Raman and Rayleigh scattering diagnostic capability in order to provide simultaneous time-resolved major species, temperature, and mixture fraction measurements in turbulent nonpremixed flames for the first time. It is intended that data produced from the newly-developed measurement capability can provide the turbulent combustion community with new insights into the time-varying effects of the turbulent
flow field on processes such as reactant mixing, ignition, extinction, flame stabilization, and pollutant formation under nonpremixed combustion conditions. In addition, just as previous low-repetition-rate Raman/Rayleigh scattering measurements have provided excellent data for comparison with time-averaged modeling approaches [6]–[9], time-resolved data will provide computational researchers with new experimental databases for assessing and validating more advanced, time-dependent modeling approaches (such as LES).

1.2 Scope of Research

Turbulent flows are highly complex phenomena which fluctuate in both space and time. When turbulent flows are coupled with chemical reactions, they become increasingly complicated, occurring over a broader range of length and time scales. In this manner, measurements that resolve flow field properties in both space and time are highly desired. Nonintrusive laser diagnostic approaches (further discussed in Section 2.3) offer the opportunity to measure the flow field velocity, temperature, and species concentrations directly without perturbing the processes of interest. Furthermore, laser-based measurements are possible with spatial resolution that is predominately determined by signal level and optical considerations rather than physical hardware constraints such as those associated with using thermocouples or other physical probes [10]. Due to the spatially- and temporally-intermittent nature of turbulent combustion processes, recent work in the field of combustion diagnostics has focused on the extension of laser-based imaging techniques to multi-kHz acquisition rates [11]–[20].
This research focuses on simultaneous temporally-resolved measurements of temperature and major species concentrations (O\textsubscript{2}, N\textsubscript{2}, H\textsubscript{2}O, and H\textsubscript{2}) measurements in hydrogen-based turbulent nonpremixed flames through the development of new, nonintrusive, high-speed combined 1D Rayleigh/Raman spectroscopy at multi-kHz acquisition rates. The simultaneous measurement of all major species allows the deduction of the mixture fraction, which describes the state of mixing between fuel, oxidizer, and products. The mixture fraction, in particular, has been identified as being important in characterizing the time-varying effects of turbulence on species transport, mixing, and the subsequent flame chemistry [21]. Since turbulent processes can be considered largely stochastic, a better understanding of the coupling between turbulence, scalar transport, and reaction chemistry is achieved through “real time” monitoring of the individual scalars and their mutual interactions. It is in this regard this research focuses on (1) the development of quantitative, simultaneous high-speed Rayleigh/Raman scattering imaging diagnostics to track temperature and major species concentrations in time and space and (2) the application of the newly-developed measurement capability to provide new physical insight into flame dynamics and the effects of fluctuating turbulence on combustion processes through the determination of integral time scales of important flow scalars. Specifically this research seeks to address the following research questions:

1. What are the accuracy, precision, and detection limits of the new multi-kilohertz Rayleigh/Raman measurements? How do these compare with conventional low-repetition rate (~10 Hz) measurements as reported
within the literature? Is the signal-to-noise ratio (SNR) sufficient to determine pertinent time scales and fluctuations? Is it possible to determine scalar gradients and their temporal evolution?

2. How do the species concentrations and temperature fluctuate in time? What are the integral time scales for the temperature, major species, and mixture fraction? Are the integral time scales equivalent for all scalars? How do they scale as a function of spatial position and Reynolds number?

3. How are the individual scalar (temperature, major species concentration, and mixture fraction) fluctuations correlated in space and time with one another? How do the correlations vary as a function of spatial position and Reynolds number?

1.3 Development of High-Speed 1D Raman/Rayleigh Imaging

The primary goal of this research is the development a new high-speed laser diagnostic capability for measuring the local temperature and major species concentration distributions in both space and time under turbulent flame conditions. Initial work in our laboratory at Ohio State demonstrated the ability to capture ten sequential, 10-kHz 1D Raman/Rayleigh scattering images, although no effort was made to place the Raman/Rayleigh signals on an absolute scale and determine temperature and species concentrations [22]. In this dissertation, the previous work is extended to produce time-resolved temperature and major species concentration (i.e., \( N_2, O_2, H_2, \) and \( H_2O) \) profiles
through detailed calibration, the implementation of a rigorous data reduction approach, and the utilization of the new High-Energy Pulse Burst Laser System (HEPBLs) [23], which is capable of providing a burst of temporally-sequential pulses with individual pulse energies of up to 1 Joule at 532 nm for durations exceeding 20 ms (>200 temporally-sequential images at 10 kHz). Specific attention is given to addressing the challenges that are particular to the high-speed 1D Raman/Rayleigh measurements; namely the complications that arise from the intersection of low signal processes such as Raman scattering and high-noise (low dynamic range), high speed imaging hardware (laser + detector system). Detailed characterization of single-shot, inter-pulse burst, and burst-to-burst accuracy and precision is presented through measurements in near-adiabatic laminar flames over a broad range of operating conditions and statistical comparisons to existing low-repetition-rate data in a set of target turbulent nonpremixed flames.

1.4 Characterization of Turbulence-Chemistry Interaction

A second objective of this research is the application of the new high-speed Rayleigh/Raman scattering imaging to investigate the time-varying structure and dynamics of turbulent nonpremixed flames. Important turbulent combustion processes occur over a wide range of spatial and temporal scales spanning from the size of the combustor to the finest turbulent scales where turbulent energy is dissipated through viscosity. These processes, which include turbulent mixing, the formation of chemical species, heat transfer, and molecular and turbulent transport, are coupled to one another
thus influencing the flame structure, combustion efficiency, and chemical processes such as pollution formation. This complexity makes direct numerical simulation (DNS) of realistic combustion environments (or even high-Reynolds number laboratory-scale flames) impossible using current computing technology. While significant progress has been made towards developing modeling capabilities such as LES, turbulent combustion models are still in development and currently are not reliable enough for predictive use. Thus, accurate measurements are required to investigate the governing chemical and physical processes occurring under turbulent combustion conditions.

In this research, the temporally-fluctuating radial profiles of temperature, major species concentrations, and mixture fraction were measured at multiple axial locations within a series of well-characterized hydrogen-based nonpremixed jet flames (DLR flames H3 and H5) [9], [24] to examine the spatially- and temporally- intermittent flame structure quantitatively. While there have been numerous qualitative “visualization” studies using high-speed imaging of flame markers such as the OH radical (see [11], [25], [26] and discussion in Section 2.5) or quantitative kHz-rate measurements of a single scalar such as temperature (for examples see [14], [20], [27]), these results yield an incomplete representation of the effects of turbulence on flame structure. Alternatively, the simultaneous measurement of temperature, the major species concentrations, and mixture fraction allows for a more complete and better understanding of chemistry-turbulence interaction within nonpremixed flames by allowing direct observation of fuel/air/product mixing, conversion of reactants to products, and the governing time scales of multiple scalar quantities.
In addition to the direct visualization of the temporal nature of the turbulent flame structure, the multi-kHz measurements yield unique set of joint spatial and temporal statistics, some of which have not been available previously, including space-time correlations, integral time scales, and multi-scalar, temporal cross-correlations. These temporally-based statistics of species, mixture fraction, and temperature give new insight into the interaction between turbulent flow (i.e. “eddies”) and scalar formation and transport. Furthermore, the determination of scalar-specific time scales has important implications for turbulent combustion modeling. Often, individual species time scales are assumed to be identical, although a numerical study by Hawkes et al. [28] suggests individual scalar timescales have a significant dependence on Schmidt number and chemical reactions. While previous measurements of temperature [14], [29] and minor species [30]–[33] time scales are available, this research is the first to measure the integral time scales of temperature, major species, and mixture fraction, collectively.

1.5 Dissertation Outline

The primary focus of this work is on technique development and validation of the high-speed combined 1D Raman and Rayleigh scattering approach. To this end, a significant portion of this dissertation is focused on characterization of the imaging system, its implementation within turbulent combustion environments, and measurement uncertainty. Chapter 2 provides background information on important combustion scalars, relevant turbulent length and time scales, and an overview of existing measurement techniques. Chapter 3 describes the current experimental methodology and
hardware configuration for the combined high-speed Raman/Rayleigh imaging system in detail. Chapter 4 discusses the data processing and calibration techniques, with specific focus on details unique to achieving quantitative major species and temperature measurements via Raman/Rayleigh scattering with current high-speed imaging hardware. Chapter 5 focuses on assessing the accuracy and precision of the high-speed measurements through measurements in near-adiabatic, laminar flames and corresponding comparisons to adiabatic flame calculations. Chapter 6 focuses on the demonstration of single-shot measurement capabilities in turbulent flames. The precision, accuracy, and detection limits of in-flame measurements using the high-speed imaging system are quantified through comparisons with existing turbulent flame data available through the TNF Workshop [24]. Chapter 7 highlights the utility of the high-speed Raman/Rayleigh measurements through direct visualization of time-dependent behavior of the scalar profiles and new space-time statistics including temporal autocorrelations, cross-correlations, and their associated integral and scalar-interaction timescales. Finally, Chapter 8 offers a summary of the main research contributions, concluding remarks, and suggestions for future work.
Chapter 2: Background

2.1 Mixture Fraction

Combustion is classified as “nonpremixed” when the fuel and oxidizer originate from separate streams. Under these conditions the local mixing (e.g., between fuel and air) controls the chemical reaction rates and is the limiting step of the combustion process.

For nonpremixed combustion, the mixture fraction $\xi$ is an important scalar, as it characterizes the extent of local molecular mixing and is defined (in general terms) as the ratio of the total mass of conserved scalar in the measurement volume to the mass of the conserved scalar originating from the fuel stream [34]:

$$\xi = \frac{\beta - \beta_{\text{oxidizer}}}{\beta_{\text{fuel}} - \beta_{\text{oxidizer}}}$$  \hspace{1cm} \text{Equation 2.1}

In Equation 2.1, $\beta$ is any conserved scalar, $\beta_{\text{fuel}}$ is the value of the conserved scalar originating in the fuel stream, and $\beta_{\text{oxidizer}}$ is the value of the conserved scalar originating in the oxidizer stream. Note that the mixture fraction is equal to 1 in the fuel stream, 0 in the oxidizer stream, and any value between 0 and 1 represents some level of mixing between fuel, oxidizer, and combustion products. In addition to being a descriptor for the local state of mixing, for many nonpremixed combustion modeling approaches, the complete instantaneous thermo-chemical state of the fluid is parameterized as a function of the local mixture fraction [35].
Specific definitions of mixture fraction depend on how preferential molecular (or “differential”) diffusion is handled and are only equivalent when all species have equal diffusivities. The TNF workshop [24] uses a definition for $\xi$ based on atomic mass given by Bilger et al [36] such that:

$$
\xi = \frac{2(Y_C - Y_{C,2})/w_C + (Y_H - Y_{H,2})/2w_H + (Y_O - Y_{O,2})/w_O}{2(Y_{C,1} - Y_{C,2})/w_C + (Y_{H,1} - Y_{H,2})/2w_H + (Y_{O,1} - Y_{O,2})/w_O} \quad \text{Equation 2.2}
$$

where $Y_C$, $Y_H$, and $Y_O$ are the elemental mass fractions of carbon, hydrogen, and oxygen respectively, $w_C$, $w_H$, and $w_O$ are the atomic weights of carbon, hydrogen, and oxygen respectively, the subscript 1 refers to mass originating in the fuel stream, and the subscript 2 refers to mass originating in the oxidizer stream. Since the mixture fraction is based on elemental mass fractions, it is most accurately obtained when simultaneous measurements of all major species concentrations are available\(^1\) [7]. The definition presented in Equation 2.2 can be modified in various ways to accommodate available data, such as excluding the oxygen terms when measurement noise and interference effects are an issue [37], [8]. As current research focuses only on hydrogen flames, a modified version of the Bilger mixture fraction is used as shown in Equation 2.3, with all carbon-containing species removed.

$$
\xi = \frac{(Y_H - Y_{H,2})/2w_H + (Y_O - Y_{O,2})/w_O}{(Y_{H,1} - Y_{H,2})/2w_H + (Y_{O,1} - Y_{O,2})/w_O} \quad \text{Equation 2.3}
$$

\(^1\) The influence of combustion intermediates and minor species on $\xi$ typically is negligible as the major species account for more than 98% of the atomic mass when simple fuels such as hydrogen or methane are used.
2.2 Relevant Length and Time Scales

An important characteristic of turbulent flows, including reacting flows, is that they contain a broad range of length and time scales. The largest characteristic length scale in the flow (referred to as the “outer length scale”), $L$, can represent the size of the flow system itself and provides an upper bound for coherent turbulent motion or eddy size. For example, $L$ can be equal to the width of a reacting jet or length of the combustor in an engine. The analogous local outer time scale can be calculated from the bulk flow velocity $U(x)$ and the outer length scale $L(x)$ as shown in [38]:

$$\tau_L = \frac{L(x)}{U(x)}$$

Equation 2.4

where $x$ describes a principle flow direction. Smaller than the outer length and time scales are the integral length and time scales, $l_0$ and $\tau$, which characterize the average size or duration of large-scale eddies in the flow [34]. The integral scales are a measure of “flow memory” or more precisely, represent the length and time it takes for a flow property to become uncorrelated with itself [39]. Kinetic energy that is extracted from the mean flow and input into the integral scales eventually “cascades” from the larger scales to the smallest fluid length scale, the Kolmogorov scale $\eta$, where it is dissipated through viscous motion into heat.

$$\eta = \left(\frac{\nu^3}{\langle \varepsilon \rangle}\right)^{1/4}$$

Equation 2.5

In Equation 2.5, $\nu$ is the kinematic viscosity and $\langle \varepsilon \rangle$ is the mean rate of kinetic energy dissipation [34], [37], [38]. By setting the rate of energy transfer from the largest eddies ($\varepsilon \sim u_0^2/\tau$) equal to the rate at which the small scales receive the energy and assuming the
local Reynolds number to be unity at the smallest length scales (such that viscous effects dominate), the ratio of the Kolmogorov scale to the outer scales can be related to the outer-scale Reynolds number for the flow [38], [39].

\[ \frac{\eta}{L} \sim Re_L^{-3/4} \quad \text{Equation 2.6} \]

\[ \frac{\tau \eta}{\tau} \sim Re_L^{-1/2} \quad \text{Equation 2.7} \]

Equations 2.6 and 2.7 show the large dynamic range in both length and time scales that can exist within turbulent flows, especially at high-Reynolds number conditions. While the Kolmogorov scale characterizes the smallest length scale in a turbulent flow or more precisely, the smallest length scale at which velocity fluctuations occur, the Batchelor scale \( (\lambda_B) \) describes the smallest length scale of relevance for scalars and was defined based on dimensional reasoning as [40]:

\[ \lambda_B = \eta Sc^{-1/2} = \left( \frac{\nu^3}{\langle \varepsilon \rangle} \right)^{1/4} Sc^{-1/2} \quad \text{Equation 2.8} \]

where \( D \) is the diffusion coefficient and \( Sc = \nu/D \) is the Schmidt number [37]. The Batchelor scale represents the smallest length scale over which a turbulent scalar fluctuation can be sustained or equivalently, a cutoff length scale [37]. Using Equation 2.6, the Batchelor scale can be written in terms of outer-scale variables as:

\[ \frac{\lambda_B}{L} \sim Re_L^{-3/4} Sc^{-1/2} \quad \text{Equation 2.9} \]

While these scaling relations were formulated for non-reacting flows, results published by Wang et al. [37] show comparable energy cascade spectra in reacting flows as

---

\(^2\) As an example, for a typical laboratory flame with \( Re_L = 10,000, \frac{\eta}{L} \sim 10^{-3} \) and \( \frac{\tau \eta}{\tau} \sim 10^{-2} \).
compared to non-reacting turbulent flows which suggest that the scaling relations also are applicable to turbulent flames.

2.3 Laser Diagnostics

Nonintrusive laser diagnostic measurements have many advantages over conventional probes\(^3\) that make them ideal for investigating turbulent flow and combustion properties including velocity, temperature, and species concentrations. Unlike intrusive measurements (i.e., thermocouples, hot-wires, or gas sampling probes), laser diagnostic approaches provide the capability for measuring flow field velocity and scalars without physically disturbing the flow, and in the case of combustion environments, without concern of probe survival at high temperatures. The absence of a physical probe also increases the potential measurement resolution by making it dependent on signal and optical constraints rather than probe size [10]; although it must be noted that very small physical probes may be preferable when optical diffraction limitations prohibit resolution of the smallest scales as in the case of high-speed flows. The signal strength in many laser diagnostic methods (including spontaneous Raman and Rayleigh scattering as discussed in Section 2.4) is proportional to input laser energy, meaning higher spatial resolution often is achieved by simply using a more powerful laser source. Furthermore, instantaneous laser-based measurements utilizing short-pulsed lasers also offer excellent temporal resolution by “freezing” the flow field over a period of the laser pulse (ns), which is much smaller than the characteristic time scales in the flow (ms) [10].

\(^3\) Here the term “probe” refers to any measurement device physically inserted into the flow field.
Another distinct advantage of using laser diagnostics over conventional probes is the ability to measure flow scalars in situ. In general, when using conventional probes, the physical properties of the probe and their variation within a flowfield are measured and then related back to specific flow properties. Alternatively, laser-based techniques involve the direct interaction between light and flow-field properties, thus increasing accuracy and measurement resolution. In addition, the nature of laser-based measurements allows researchers to isolate specific properties (i.e., velocity, temperature, or individual species) for interrogation through careful selection of laser wavelength and signal detection.

Generally, laser diagnostic approaches are applicable only when optical access to the flow field is available. These constraints frequently preclude their use in realistic combustion environments, where enclosed (windows or walls) and/or sooty environments are found. Mie scattering from soot particles often overwhelms other signals and the particles themselves collect on optical windows, reducing optical access. These issues are generally less important for experiments aimed at understanding specific turbulence or combustion phenomena and those targeted towards model-validation experiments, as the combustion geometries and flame properties can be carefully chosen to avoid these issues. The measurements presented in this dissertation are conducted within a series of canonical turbulent nonpremixed flames previously identified within the TNF Workshop [24] to optimize the applicability of laser diagnostics for understanding turbulence-chemistry interaction and model validation [7].
Finally it is noted that there are many comprehensive textbooks [10], [41] and review papers [7], [42] outlining the methodology, implementation, attributes, and shortcomings of specific laser-diagnostic approaches in combustion environments. Generally speaking, measurements target specific state descriptions of the flow including kinematic (velocity), mixing (mixture fraction), chemical (major and minor species; soot properties), and thermodynamic (temperature, pressure, and density) properties. While a comprehensive review of the most common laser diagnostic methods is left for aforementioned texts [7], [10], [41], [42], a brief review of the theory for spontaneous Raman and Rayleigh scattering is presented below in Section 2.4.

### 2.4 Raman and Rayleigh Scattering

Turbulent flames contain many different scalars, each of which can play a vital role in various combustion processes and have significant effects on flame stability, combustion efficiency, or pollutant formation. As described previously, the majority of laser diagnostic techniques are property-specific, thus it is often beneficial to combine multiple diagnostic techniques to simultaneously measure multiple scalars for a more complete description of the local thermo-chemical state of the flow. By combining spontaneous Rayleigh and Raman scattering, both the temperature and concentration of the major species can be deduced [43]. This section provides a brief background on the principles governing Rayleigh and Raman scattering and its implementation in combustion environments via laser-based excitation.
Figure 2.1: Energy levels diagram for Rayleigh and Raman transitions. Virtual states can be thought of as “the ‘wings’ of the probability function of nearby states” [44].

When the electro-magnetic field from a laser interacts with atoms or molecules, electro-magnetic waves may be re-emitted, which depending on the details of the interaction process can be characterized as scattering or fluorescence. If the atom or molecule is excited into a virtual energy state that “instantaneously” decays back to its original energy state by emitting radiation at the same frequency as the incident (oscillating) electric field of the laser, the process is termed elastic scattering (i.e., the wavelength of the emitted light and incident photons are equal). For particles much smaller than the wavelength of the incident photon, this phenomenon is called Rayleigh scattering. Alternatively, Raman scattering describes the inelastic scattering of light, where an incident photon from the laser excites the atom or molecule into a short-lived virtual energy state that decays to a different energy state. If the incident photon loses energy to the molecule, then emission occurs at a lower frequency (termed Stokes-shifted Raman scattering) and when the photon gains energy from the molecule, emission occurs
at a higher frequency (termed anti-Stokes-shifted Raman scattering). A schematic of these processes is shown in Figure 2.1.

Rayleigh and Raman scattering are considered to be virtually instantaneous because the existence of an electron in these states is very improbable, leading to lifetimes < $10^{-12}$ seconds [10]. The frequency shift of the Stokes and anti-Stokes scattered photons are given as:

Stokes frequency shift \[ \nu_S = \nu_0 - \Delta \nu \quad \text{Equation 2.10} \]

Anti-Stokes frequency shift \[ \nu_{AS} = \nu_0 + \Delta \nu \quad \text{Equation 2.11} \]

where $\Delta \nu$ is the difference between the upper and lower energy states for the transition as shown in Figure 2.1. It is seen from Equations 2.10 and 2.11 that the frequency shift is independent of the frequency of the incident laser, meaning that for a given species, the frequency shift in wavenumbers is fixed from the incident laser (and Rayleigh) frequency (wavelength). The magnitude of this shift is dependent upon the energy gained or lost in the molecular bond, making the frequency shifts species specific, at least for small molecules. This feature is exploited in combustion environments to measure the concentration of specific components (typically the major species) contained within a gas mixture. Equations 2.10 and 2.11 also imply that the Raman effect is a non-resonant process which can occur for any frequency of incident laser light, thus negating the need for frequency-tunable laser sources and greatly simplifying the experimental approach.

Equations for the Rayleigh and Raman signal intensities are given as:

\[
I_{Ray} = C_{Ray} I_0 N \frac{d\sigma_{Ray}}{d\Omega} \Omega L \quad \text{Equation 2.12}
\]
\[ I_{\text{Ram},i} = C_{\text{Ram},i} I_0 N_i \frac{d\sigma_{\text{Ram},i}}{d\Omega} \Omega L \]  

Equation 2.13

where C is a calibration factor based on the experimental setup, \( I_0 \) is the incident laser intensity, \( N \) is the number density, \( d\sigma/d\Omega \) is the differential scattering cross section\(^4\), \( \Omega \) is the collection solid angle, and \( L \) is the length of the probe volume [41]. Rayleigh and Raman signals are weak compared to other common laser diagnostic techniques as the associated cross-sections are small, on the order of \( 10^{-28} \) [45] and \( 10^{-31} \) cm\(^2\)/sr for Rayleigh and Raman scattering, respectively [10]. A table of cross-sections for the combustion species considered in this research is provided in Table 2.1. Raman and Rayleigh cross-sections display a \( \lambda^{-4} \) dependence with respect to laser wavelength, meaning they increase rapidly with decreasing wavelength. While this may suggest that UV light should be used for Rayleigh and Raman scattering diagnostics, there is an increased probability of fluorescence interferences in this range. In addition, the calibration factor C also is dependent on wavelength due to varying efficiencies in available collection optics and detectors. Researchers typically take advantage of the high-powered energies available from the visible output of Nd:YAG lasers and the high collection efficiency of visible optics and detector technology by using 532-nm light for combined Raman/Rayleigh experiments [37], [8], [10], [41], [46]–[48], [9]. In addition, Masri et al. [49] showed that interferences in hydrocarbon flames are minimized in the 500 – 600-nm wavelength range, which coincides with the 532-nm output from the frequency-doubled Nd:YAG laser.

\(^4\) For Rayleigh scattering, the differential scattering cross section is effectively a “mixture-averaged” differential scattering cross section which is calculated as \( d\sigma_{\text{Ray}}/d\Omega = \sum_i X_i d\sigma_i/d\Omega \), where \( X_i \) and \( d\sigma_i/d\Omega \) are the mole fraction and differential scattering cross section for species i, respectively.
Species | Raman cross-section at 532 nm (10\(^{-30}\) cm\(^2\)/sr) [10] | Rayleigh cross-section at 532 nm (10\(^{-28}\) cm\(^2\)/sr) [45]
---|---|---
N\(_2\) | 0.46 | 6.24
H\(_2\) | 0.943 | 1.3
O\(_2\) | 0.65 | 5.4
H\(_2\)O | 0.9 | 4.4

Table 2.1: Species scattering cross-sections at 295 K

As shown in Equations 2.12 and 2.13, the Rayleigh signal intensity is linearly proportional to the total number density within the probe volume and the mixture-averaged Rayleigh scattering cross-section, \(d\sigma_{\text{RAY}}/d\Omega\), which is calculated as the summation of each individual species cross-section weighted by its mole fraction. In contrast, the Raman signal intensity is linearly proportional only to the number density and Raman scattering cross section of the individual species of interest [41]. The different signal dependences of Raman and Rayleigh scattering allow the deduction of the local temperature and number density of major combustion species when the two scattering processes are measured simultaneously. This Raman/Rayleigh approach has been applied successfully at low repetition rates within the combustion community for quite some time as described below and is the targeted approach of this dissertation research.

2.5 Applications of Raman and Rayleigh Scattering in Combustion Environments

While spontaneous Raman scattering (SRS) was often used for time-averaged measurements within non-reacting flows, the strength of background flame luminosity compared to weak Raman signals precluded its use in most combustion environments...
[50] until the late 1970s and early 1980s, where gated signal collection via boxcar integrators [51]–[54] minimized background luminosity to acceptable levels to utilize collected Raman signals. In addition, pulsed laser sources became readily available and enabled major species and temperature measurements via pulsed SRS in turbulent combustion environments [43], [55]–[57] due to higher signal-to-background levels afforded by the pulsed laser sources. Though any pulsed laser source can be used for spontaneous Raman scattering, pulse strength, potential interferences, and available optics have limited previous SRS measurements to three main types of pulsed laser sources [58]: flash-pumped dye lasers [43], [56], [59], UV excimer lasers [60], [61], and frequency-doubled Nd:YAG lasers [62]–[65]. A previous study by Meijer and Heck [66] compared signal levels to background interference and determined the 532-nm output from pulsed Nd:YAG lasers to be the most suitable wavelength for SRS measurements. This, combined with high pulse energies make Nd:YAG lasers an ideal choice for most SRS experiments [66].

In general, there have been two detection approaches for previous spontaneous Raman scattering measurements: photomultiplier tubes (PMTs) or array detectors such as CCD cameras. Early spontaneous Raman scattering setups utilized multiple PMTs for single-point measurements, where each species-specific Raman signal was collected on an individual PMT. While these systems (see examples in [43], [53], [67]) were somewhat cumbersome and required numerous calibrations, they minimized dynamic range issues by allowing for the gain on each “species channel” (in this case a separate PMT) to be adjusted individually. In contrast, array detectors (such as intensified-CCD,
CCD, or – in this study – CMOS cameras) allow for more complete spectral information and the interrogation of spatial structures via one-dimensional “line imaging” [41]. Groups at Sandia National Laboratories [37], [46], [47], [6], [68], TU Darmstadt [64], [69], and others [70]–[72] have used 1D SRS to interrogate turbulent mixing and turbulent-chemistry interaction in turbulent non-premixed and premixed flames, where the one-dimensional aspect of the measurement allows the deduction of relevant spatial scales and a 1D surrogate of the scalar dissipation rate, which describes the rate of mixing.

Spontaneous Raman scattering is rarely used on its own. While early SRS measurements used spectral fitting to nitrogen lines for temperature measurements, more recent studies rely on a simultaneous spontaneous Rayleigh scattering measurement for deducing temperature[8], [43], [73]. Raman scattering also is commonly combined with laser-induced fluorescence (LIF) in order to obtain measurements of minor species concentrations [60], [63], [6], [74], [75]. These types of multi-scalar measurements have been invaluable within the context of the TNF Workshop [24], as they provide information for assessing combustion models [41].

More recently, there has been a great deal of focus on extending SRS measurements to increasingly complex fuels and more complicated combustion environments. The majority of Raman/Rayleigh scattering flame measurements have been limited to hydrogen- and methane-fueled flames in simple, unconfined, atmospheric flames, though very recent work at the CRF has extended extending Raman/Rayleigh/LIF measurements to oxygenated fuels such as dimethyl ether [76]–[78] and “oxyfuel”
environments [79]. More advanced combustion environments such as high pressure [80]–[83], jet-in-cross flows [84], swirl burners [85]–[87], and model gas turbine combustors [82], [83], [86], [88] also have been investigated by researchers at Sandia National Laboratories, Technical University of Darmstadt, and the German Aerospace Center (DLR).

2.6 High Speed Imaging

Much of the understanding of turbulent combustion in recent years has come from the emergence of non-intrusive laser-based measurements of the flow field properties. Since turbulent flows are characterized by unsteady and quasi-stochastic behavior, spatially- and temporally-correlated laser-based measurements are highly desired in order to capture the dynamic nature of important processes occurring in turbulent combustion environments including reactant mixing, flame stabilization, ignition/extinction, and instabilities resulting from the coupling of acoustic, flow, and chemical processes. This requirement dictates that velocity and scalar measurements should be acquired at acquisition rates faster than the turbulent time scales of interest. For laboratory-scale flows, the relevant time scales typically are less than 1 ms, thus acquisition rates > 1 kHz are required. In addition, time-resolved measurements allow the identification of combustion events whose occurrence may be rare, but extremely important including engine misfire, flashback, local extinction, and flame blowoff or blowout. However, many gas-phase scalar measurements require high-energy excitation sources such as

---

5 See Section 3.6 for a discussion of expected time scales and sampling requirements.
excimer or Nd:YAG lasers, which traditionally have been limited to low repetition rates on the order of 10 to 100 Hz. Only recently have advances in high-speed laser systems and camera technology made the collection of temporally-resolved data possible. For in-depth overviews on the progression of high-speed measurement techniques, the reader is referred to [25], [26], [11].

The typical approach to high-speed imaging is the use of commercially-available, diode-pumped, solid-state (DPSS) lasers and CMOS-based camera systems. In the non-reacting fluid mechanics community, the most common use of DPSS lasers and CMOS cameras is high-speed particle image velocimetry (PIV), while for combustion studies, PIV and OH planar laser-induced fluorescence is common (see [26] and [44] for a detailed listing). Combustion studies often couple PIV measurements with planar laser-induced fluorescence (PLIF) of minor species such as OH in order to examine the effect of the turbulent velocity field on transient combustion phenomena such as extinction [12], [89], flashback [90], [91], and local extinction/re-ignition [92], [93]. Other examples of high-speed imaging using DPSS lasers include tracer-LIF measurements to image fuel-air ratios in non-firing engines (e.g., [94]–[98]), mixture fraction in unsteady, non-reacting jets [99], and temperature distributions near surfaces [100], respectively; laser-induced incandescence (LII) measurements for time-resolved soot distributions in flames [101]; 1D Rayleigh scattering-based thermometry in nonpremixed flames [20]; NO distributions in a plasma torch [102]; high-speed phosphor-based thermometry [103]; and CH distributions in lifted nonpremixed flames to identify reaction zone dynamics [104].
While many advances have been made in high-speed imaging approaches as outlined above, many methods commonly used at low repetition rates in combustion environments are difficult using continuously-operating DPSS lasers due to relatively low output energies at high repetition rates. For example, spontaneous Raman scattering, planar Rayleigh scattering and PLIF imaging of several radicals and intermediates (i.e., CO, CH\textsubscript{2}O, O, H, etc.) have either not been performed with DPSS lasers or have been sparsely demonstrated with low SNR. As noted in the recent review from Thurow et al. [6], pulse burst laser systems allow the generation of a series of temporally-sequential, high-energy laser pulses over a limited period. Because of the low duty cycle of the pulse burst laser systems, the time-averaged power output is low enough to avoid system damage (i.e., below thermal damage threshold of the solid-state gain medium) while producing very high-energy pulses over the duration of the burst. Pulse burst technology is used in the current research to facilitate the development of the simultaneous high-speed 1D Raman/Rayleigh imaging in turbulent flames through the use of the high-energy pulse burst laser system (HEPBLS) [23], [105], [29]. While the HEPBLS and its system characteristics will be treated separately in Chapter 3, it is noted that pulse burst systems have been used to perform MHz-rate NO and NO\textsubscript{2} PLIF imaging in high-speed flows [106]–[109], ultra-high-framing rate OH PLIF imaging in sonic hydrogen jet flames [110], kHz-rate spontaneous Raman scattering in non-reacting jets [65], kHz-rate planar Rayleigh scattering imaging in turbulent jets and flames [105], [29], [19], [27], and kHz-rate CH [111] and CH\textsubscript{2}O PLIF [112]–[114] imaging in turbulent flames.
As will be described in Chapter 6, one of the true utilities of the current measurements is the derivation of integral time scales of the temperature, major species, and mixture fraction. To date, very few scalar time scale measurements have been reported in the literature. Integral time scales of temperature [9, 15] and OH [31], [115] have been reported in turbulent nonpremixed flames, but there are no previously-reported time scales on major species, mixture fraction, or multiple scalars simultaneously to directly examine their inter-dependency. Very recently, high-speed point-based spontaneous Raman scattering (SRS) was demonstrated at 1 kHz in unsteady laminar hydrocarbon flames by NASA Glenn researchers utilizing a commercial diode-pumped Nd:YLF laser and intensified CCD camera in subframe burst gating mode to investigate low-frequency oscillations in laminar flames [116], [117]. While temporal auto-correlation and cross-correlation functions were presented for two species, the 1-kHz sampling rate (as limited by SNR) is unsuitable for turbulent flame studies. In the current results reported in Chapter 6, temporal auto-correlation and cross-correlation functions are presented from 1D 10-kHz measurements of all major species, which is a sufficient sampling rate to measure scalar integral time scales for the turbulent flames investigated\(^6\).

\(^6\) Based on previous studies of OH time scales in the DLR H3 flame [31].
Chapter 3: Experimental Methods

The primary goal of this research is to develop a new laser diagnostic approach capable of providing physical insight into the effects of turbulence on species transport and combustion chemistry through temporally-correlated simultaneous 1D Raman and Rayleigh scattering imaging. The Raman and Rayleigh scattering imaging yields 1D measurements of temperature and major combustion species concentrations. Of particular interest is the deduction of the mixture fraction from the major combustion species measurements, as the mixture fraction uniquely describes the state of turbulent mixing.

The experimental measurements discussed in this dissertation were developed and acquired in the Turbulence and Combustion Research Laboratory (TCRL) at Ohio State University. The experimental setup consists of a combined 1D Raman and Rayleigh line imaging system utilizing “high-speed” detection at a 10-kHz sampling rate, as first described in [22], and shown in Figures 3.1 and 3.2. The OSU Raman/Rayleigh imaging system was inspired by a similar system developed at the Combustion Research Facility (CRF) at Sandia National Laboratories [37] for data acquisition at low repetition rates, although there are many differences dictated by the uniqueness of the current high-speed measurements. The most notable difference is the use of high-framing-rate CMOS cameras and a high-speed IRO in lieu of low-noise CCD cameras in order to achieve 10-kHz acquisition rates. A second significant difference is the use with the High-Energy
Pulse Burst Laser System (HEPBLS) [23] as the excitation source as compared to a series of conventional 10-Hz Nd:YAG lasers. The particular attributes of the high-speed imaging system are discussed in detail in this chapter and the corresponding ramifications of high-speed acquisition on Raman/Rayleigh imaging (in terms of signal-to-noise, accuracy, precision) is discussed in detail in subsequent chapters.

Figure 3.1: Combined 1D Raman/Rayleigh scattering imaging setup in the Turbulence and Combustion Research Laboratory at Ohio State University. System detects simultaneous one-dimensional measurements of major species (O$_2$, N$_2$, H$_2$O, and H$_2$) and temperature using excitation from the second harmonic of an Nd:YAG laser source (532 nm).
Figure 3.2: Photographs of the 1D Raman/Rayleigh scattering setup. (Top) View of the laser path through the test section (green line) as it passes through an alignment iris, over the McKenna burner, and over the turbulent jet-in-coflow burner. (Bottom) The scattered light was collimated and focused by the achromats and arranged in a periscopic configuration into the spectrometer by a set of mirrors.

3.1 Signal Collection

The Raman and Rayleigh scattering system (shown in Figure 3.1) used in this work is based on the excitation from frequency-doubled output of Nd:YAG laser systems and the subsequent collection of the Rayleigh- and Raman-scattered light into an imaging spectrometer. Two laser excitation sources were used as described below: (1) a “conventional” pulsed Q-switched Nd:YAG laser operating at 10 Hz and (2) a custom
high-energy pulse burst laser system (HEPBLs) operating at 10 kHz. For either laser system, the 532-nm laser output is focused into the probe volume within a jet-in-coflow facility (or above calibration burners) by a single 750-mm plano-convex spherical lens (anti-reflective coated on a fused-silica substrate). The lens, while fixed in the direction of the laser propagation axis, can be adjusted in the vertical direction by a motorized linear actuator (Newport NSA12) in order to control beam stability throughout the testing. The converging beam passes through an anti-reflective coated fused-silica window at a 45 degree angle to the beam path. This window allows a small portion (<0.25%) of the focusing beam to be reflected and re-directed towards a CCD camera (for pulse-stretcher alignment) or a high-speed energy meter as shown in Figure 3.1.

After transmitting through the window, the excitation beam first passes above the surface of a McKenna burner [59] operating with filtered air. A high-speed CMOS camera (Phantom v711) was placed in front of the McKenna burner with a 20-mm spacer and 50-mm focal length, f/1.2 Nikon lens for Rayleigh scattering measurements in the uniform stream of air. These measurements are used for relative energy corrections from individual pulses as Rayleigh scattering fluctuations in a flow with uniform composition, pressure, and temperature are a direct result from pulse-to-pulse energy fluctuations. In order to minimize scattering within the field-of-view of the cameras, the beam travels though beam tubes both prior to and after passing the test sections, which were surrounded on three sides by black laser curtains.
Figure 3.3: Raman/Rayleigh experimental setup. (Top) A detailed schematic and optical layout of the high-throughput, imaging spectrometer. (Bottom) A photograph of the optical layout of the spectrometer. Note that in this experiment, the 532-nm band-pass filter (4) was removed and a second 532 nm notch filter was placed in the Raman optical path to ensure the removal of all Rayleigh light from the collected Raman signals.
The Rayleigh- and Raman-scattered light is collected by a pair of 150-mm-diameter achromats (Qioptic Linos Phototonics, f=2.0, 300 mm focal length and f=4.0, 600 mm focal length), yielding a magnification of 2.0. The collected signal is focused through a 13.6-mm tall, adjustable-width entrance slit (Thorlabs VA100) on a custom-built combined Raman and Rayleigh scattering spectrometer shown in Figure 3.3. The imaging spectrometer design is based off of the combined Rayleigh/Raman/CO-LIF imaging spectrometer at Sandia National Laboratories as described in [48].

Inside the spectrometer, the combined Raman- and Rayleigh-scattered light is collimated by a 105-mmm f/1.8 Nikon lens and then separated by a dichroic beam splitter (Semrock). The dichroic beam splitter acts as a longpass filter which transmits light at wavelengths longer than 548 nm (Raman scattering), while reflecting shorter wavelengths (Rayleigh scattering). The reflected Rayleigh signal (532 nm) is focused into the first camera (Phantom v710) with a Nikon 50mm, f/1.4 camera lens. A neutral density filter is placed in front of the camera lens to reduce the signal incident on the Rayleigh camera while keeping the lens fully open to minimize signal reduction near the edges of the field of view. The signal that transmits through the dichroic then passes through a 532-nm notch filter (Semrock Laser Line Filter) to block the 532-nm Rayleigh scattering and isolate the Raman signal. The remaining signal is then focused down through a 50-mm f/1.2 Nikon camera lens, re-collimated by an 85-mm, f=1.4 Nikon lens to achieve the desired magnification, and projected onto a volume phase holographic (VPH) transmission grating (Kaiser Optical, 1200 lines/mm, 22° incident/refracted angles, >85% efficiency from 550 nm to 680 nm). Between the camera lenses, the signal transmits.
through a second 532-nm notch filter to remove residual 532-nm signal. The VPH grating spectrally disperses the species-specific Raman-shifted light onto a high-speed image intensifier (LaVision HS-IRO), which is coupled to an 85-mm, f/1.4 Nikon camera lens and a high-speed CMOS camera (Vision Research, Phantom 710). This optical setup, which differs from that previously used in [22], produces a system magnification of approximately 1.0, allowing nearly 1:1 imaging on both the Raman and Rayleigh channels. Based on the CMOS camera sensor size, a system magnification of 1.0 allows one-dimensional imaging of a 13-mm line segment. Because of edge effects from the slit, data presented in this dissertation are processed over a 6.4-mm line segment.

3.2 Laser Excitation Systems

3.2.1 10-Hz Q-Switched Nd:YAG Laser

A key goal within the technique development is the characterization of the effects of high-speed imaging hardware on the collected Raman and Rayleigh signals. In order to reduce additional experimental uncertainty from the laser source, a commercially-available Q-switched Nd:YAG laser (Quanta-Ray Pro-290) operating at 10 Hz was used in a series of measurements to explicitly characterize the quality of the measurements due to the imaging hardware only. These measurements allow for the examination of optical system limits, including SNR values and measurement accuracy, under “ideal” conditions with a sharp beam focus and low pulse-to-pulse fluctuations. The maximum 532-nm output of the Nd:YAG laser was 850 mJ with a pulse duration of 8 ns, though lower energy levels were used in the measurements discussed in this dissertation. The laser
timing is controlled through a delay/pulse generator (Stanford Research Systems DG645) by triggering the flashlamps and Q-switch output. After focusing the beam into the test section, the 10-Hz beam displayed a FWHM of 120 µm (1/e^2 of 220µm) and 1% pulse-to-pulse energy fluctuations.

3.2.2 High-Energy Pulse Burst Laser System

High-speed (10 kHz) measurements were acquired with the first version of the HEPBLS as described in [23] and shown in Figures 3.4 and 3.5. An updated system is currently being finalized and may benefit future high-speed Rayleigh/Raman scattering as discussed in Chapter 8. The current HEPBLS is a master oscillator, power amplifier (MOPA) system which is designed to produce a series of high-energy pulses (up to ~1 J/pulse at 532 nm) at high repetition rates (>10 kHz) over extended burst durations (up to 25 milliseconds). Continuous output from a pulsed laser (Crystal Laser, Reno, NV) operating at 1064 nm is amplified in a series of custom flashlamp-pumped Nd:YAG amplifier stages, creating a “burst” of high-energy pulses. The master oscillator is a 150-mW, narrow linewidth (2.5 GHz) Nd:YVO₄ laser with 25-ns pulse duration capable of variable repetition rates ranging from 1 to 250 kHz. At a repetition rate of 10 kHz, the output was approximately 8 µJ per pulse. Prior to amplification, output from the pulsed oscillator is spatially filtered through a near-diffraction-limited diamond pinhole in order to create a more Gaussian-like beam (in space) and reduce diffraction effects in later stages, thus reducing the laser energy to approximately 4 µJ per pulse. The first and second amplification stages are double-pass, dual-flash-pumped amplifier configurations,
consisting of a polarizing beam splitter, 4-mm and 6.3-mm-diameter Nd:YAG rods, respectively, a quarter wave plate, and a zero-degree mirror.

Between the amplification stages, the beam passes through image relay optics consisting of a lens pair that focuses the beam through a diamond pinhole approximately ten times the diffraction-limited spot size and re-collimates the light at the magnification necessary to fill approximately 80% of the Nd:YAG rod in the next amplification stage. After the second amplification stage, the initial pulse train is amplified to output energies of approximately 150 mJ per pulse at 1064 nm. The pulses are then retro-reflected through a stimulated Brillouin scattering (SBS)-based phase conjugate mirror (PCM) [118] towards amplification stage 3. The PCM used in this system is an optical cell filled with a high refractive index fluid (FC-75). As the PCM has an energy threshold that must be reached before reflection, the PCM allows amplified spontaneous emission (ASE) and unamplified pulses to pass through before reflecting the amplified pulses, thereby decreasing unwanted signals that could limit achievable gain in subsequent amplifier stages.

The HEPBLS was designed such that the pulse train is split into two legs after the PCM via a 50/50 beamsplitter. The motivation for this design was to (1) delay gain saturation effects in order achieve ultra-high pulse energies; total energies that would be greater through the combination of two legs (A and B in Figure 3.4) rather than the amplification of a single burst train and (2) allow multi-parameter measurements where each output leg could be used for a separate scalar measurement. In the work described in this dissertation, the beam splitter shown in Figure 3.4 was replaced with a mirror.
sending all of the post-PCM energy into the A-leg of the HEPBLS. Amplification stages 3A, 4A, and 5A have 9.5-mm, 19-mm, and 19-mm diameter Nd:YAG rods respectively and are configured in a single-pass geometry. The relay imaging optics between legs 3A and 4A and 4A and 5A have vacuum tubes in order to ensure no optical breakdown occurs when the beam is focused. The output diameter of the beam after AMP 5B is reduced through the use of a Galilean telescope before the beam is frequency-doubled through a 12 mm x 12 mm x 20 mm type-I lithium triborate (LBO) crystal. The conversion rate from 1064 nm to 532 nm is approximately 50% at the pulse energies used in this study, resulting in ~20 ms, 532-nm burst trains with ~0.6-1.0 Joules/pulse at 10 kHz for selected gain settings.

Figure 3.4: Schematic of the High-Energy Pulse Burst Laser System (HEPBLs)
For the work presented here, the HEPBLS outputs 170 sequential pulses within a single burst at 10 kHz. Due to heating effects, the focus of the 10 kHz beam shifts slightly in the radial direction from the beginning to the end of the burst, causing the focus of the beam to move within the measurement volume throughout the burst. Figure 3.6 shows a 2D map of the changing beam shape through a sample pulse burst. Each vertical slice is the normalized spatial profile taken as a vertical slice from the center of the Rayleigh camera. The blank spaces on either end represent the images added at the beginning and end of the burst to provide an image background on the cameras. Figure 3.7 shows the beam profile averaged over 151 consecutive laser shots within the burst. While the instantaneous images give perspective on how the pulse changes over time, the average beam width (140 \( \mu m \) FWHM, 280 \( \mu m \) 1/e\(^2\)) serves as an upper estimate for the resolution of the measurement. In [23], the system characteristics of the HEPBLS were
described in detail, where pulse-to-pulse fluctuations within a single burst varied by less than 9% with a resultant RMS fluctuation of 5% over the burst duration. Subsequently, stability issues were noted with the Nd:YVO₄ seed laser leading to undesirable pulse-to-pulse fluctuations⁷. Unfortunately, during the data campaign responsible for the data reported within this dissertation, pulse-to-pulse energy fluctuations between 9 and 36% were observed. Two sample energy traces are shown in Figure 3.8. The effects of these energy fluctuations will be discussed in Chapter 4 in terms of SNR considerations and measurement accuracy, especially for small-concentration measurements.

Figure 3.6: Normalized spatial profile of HEPBLS output beam as a function of pulse number within a single burst. Spatial profile is determined from 1D Rayleigh scattering in air.

⁷ The Nd:YVO₄ seed laser has been replaced on the newest version of the HEPBLS. The current seed source displays RMS fluctuations of less than 1%.
Figure 3.7: Spatial profile of HEPBLS. The blue line is the spatial profile averaged over 150 sequential pulses.

Figure 3.8: Output energy as a function of burst pulse number in two sample HEPBLS pulse bursts.
3.3 Pulse Stretcher

For short-duration laser pulses, it is common to employ a laser pulse stretcher within Raman/Rayleigh scattering setups [48], [63], [64]. The motivation for this lies in the fact that Raman scattering is a very weak scattering process and is linearly proportional to the incident laser energy. Because of this, it is highly desired to use as much laser energy as possible to achieve sufficient signal. However, with high-energy lasers, the intensity of focused laser beams can exceed the threshold for dielectric breakdown of the gas in the probe volume. One way to mitigate this problem is to use the same laser pulse energy, but “temporally stretched” to reduce the laser intensity (and therefore bring it below the breakdown threshold). The utility and implementation of optical pulse stretchers has been discussed in detail by Nguyen [119]. As one example from a preliminary study [22] using a pulse-burst laser system, the energy contained in 10-ns pulses had to be limited to approximately 150 mJ in order to avoid dielectric breakdown from the focused beam. Because of this, a 3-cavity pulse stretcher similar to those described in [119] was designed in order to “stretch” the pulses temporally and therefore reduce the maximum intensity in any single pulse to avoid reaching the breakdown threshold ($\sim 3.5 \times 10^{11}$ W/cm$^2$ in air at 1 atm [10]). The pulse stretcher, shown schematically in Figure 3.9 increases the temporal profile of the beam by creating a series of right-triangle optical delay cavities into which a portion of the beam is transmitted through a beam splitter. A portion of the laser light enters into the cavity through a beam splitter while the remainder is reflected to the next cavity. The transmitted portion is sent along a delay path before passing through the other side of the beam splitter. Here the beam either
follows the same beam path as the originally-reflected portion or is again reflected to undergo another round trip through the cavity. Multiple cavities of varying length are placed in succession in order to smooth intensity peaks generated from the first cavity and further reduce the overall peak laser intensity.

Figure 3.9: A schematic diagram of the three-leg pulse stretcher

Figure 3.10: The temporal profile of the output from a 15-ns (1/e² value) 10-Hz pulse.
Each cavity used in the pulse-stretcher design described here consists of a 40/60 reflectivity/transmission beam splitter and 3-5 99.5% reflectivity Nd:YAG laser mirrors. The optical path lengths in the three cavities were approximately 8, 4, and 2 m corresponding to a temporal delay of 26.7, 13.3, and 6.7 ns for each respective cavity. Figure 3.10 shows a trace of the output pulse from an initial implementation of the pulse stretcher using a commercial, 10-Hz Q-switched Nd:YAG laser. The initial 15-ns (1/e² value) pulse is “temporally stretched” to 107 ns via a three-leg pulse stretcher. The total energy leaving the pulse stretcher was approximately 75% of the input energy and the peak laser power equal to ~10% of the original laser pulse, thus allowing the use of much higher pulse energies without breakdown.

While the pulse stretcher was employed with success in a set of preliminary measurements using the 10-Hz, Q-switched, Nd:YAG laser [65], the pulse stretcher was not used in the 10-kHz measurements presented in this dissertation for two reasons: (1) the coflowing air stream of the turbulent jet-in-coflow burner facility (see Section 3.4) was redesigned for better filtration of particulates (and therefore increase the breakdown threshold) [10] and (2) the pulse stretcher did not work as intended with the output of the current HEPBLS system. While the temporal width of the beam was increased effectively, the use of a pulse-stretcher in combination with the rapid series of pulses from a single HEPBLS burst created low-energy wings in the spatial dimension. An example of the spatial profile after implementing a single-leg pulse stretcher is shown in Figure 3.11. The example shown has a FWHM of ~140 μm and 1/e² ~360 μm (taken at ~200 mJ/pulse energies). Preliminary measurement with two optical cavities indicated
that the beam intensity distribution degraded even further when multiple optical cavities were employed. Most likely this effect is due to transient heating effects that occur throughout a single burst, leading to changes in the spatial profile of each individual pulse throughout the burst. In addition, thermal issues also lead to changes in the divergence and focusing properties of a beam that may result in undesirable beam spatial profiles for effective laser path lengths approaching hundreds of meters when using the pulse stretcher. In order to preserve the spatial resolution in the system, measurements were taken without the use of a pulse-stretcher; however as discussed below, because of the improved coflow filtration, pulse energies approaching 600 mJ could be used for the Raman/Rayleigh scattering measurements.

Figure 3.11: Rayleigh image (left) and spatial profile (right) of the 10-kHz beam after the implementation of a 1-leg pulse stretcher.
3.4 Burners and Test Facilities

3.4.1 McKenna Flat-Flame Burner

A McKenna burner (shown in Figure 3.12) [59] was used to create a uniform, dust-free, steady stream of air for relative laser-energy measurements via 1D Rayleigh scattering imaging. The 6-cm diameter honeycomb matrix burner was connected to high-pressure facility air which was filtered through a series inline compressed air filters (IAP 5 µm prefilter, 0.01 µm oil removal, and 0.01 µm activated carbon) to ensure removal of particulate. The burner is placed along the laser path prior to the test section. When used in conjunction with a high-speed camera, this configuration allows for kHz-rate measurements of pulse-to-pulse energy fluctuations. In a steady flow without temperature, pressure, or species fluctuations, fluctuations in the collected Rayleigh scattering signal corresponds to fluctuations in the laser pulse energy only. The outer surfaces of the McKenna burner and camera (shown in Figure 3.12) were covered in black cloth in order to reduce the amount of scattering from the metal/white surfaces.

Figure 3.12: Photograph of McKenna burner and energy measurement camera
3.4.2 Near-Adiabatic Hencken Burner

A Hencken burner [120] was used to create a set of well-characterized, near-adiabatic, laminar flames for use in Rayleigh and Raman system calibrations. The flames are stabilized above the surface of a 25.4 mm x 25.4 mm matrix burner, a schematic of which is shown in Figure 3.13. The burner was operated with hydrogen/air mixtures with hydrogen flow rates ranging from 2-27 SLPM, a constant air flow rate of 33.4 SLPM, and a 32.5 SLPM nitrogen coflow surrounding the flame to shield it from dust and room currents. A photograph of a $\phi = 0.7$ H$_2$/air flame is shown in Figure 3.14. All measurements were taken 18 mm above the burner surface in order to minimize heat loss and ensure near-adiabatic and near-equilibrium flames. For these calibration measurements, the turbulent jet burner was translated vertically downward and out of the optical beam path and the Hencken burner was placed into the beam path via optical rails (seen in the middle image of Figure 3.2).
Figure 3.13: Hencken Burner schematic from [120]

Figure 3.14: Image of a H₂-air Hencken flame at $\phi=0.7$. The green line represents the location of the laser 18 mm above the burner surface.
3.4.3 Turbulent Jet Flame in Co-Flow

The turbulent nonpremixed jet flames chosen for this study issue from a 7.75-mm diameter tube with a thinned rim into a 30cm x 30cm square co-flowing stream of air operating at 0.3m/s as shown in Figure 3.15. The burner is designed to give a similar flow field to those used at Sandia and the DLR [8]. Filtered, high-pressure, facility air is sent into the bottom of the coflow through two 2.54-cm-diameter tubes welded to either side of the burner. The air then flows upward through a HEPA filter, two perforated plates, 3.8 cm of honeycomb, and another perforated plate approximately 5 cm below the exit of the fuel tube. The HEPA filter is designed to eliminate any residual particles in the air, while the combination of the perforated plates and honeycomb is intended to act as “flow straighteners”, providing a nearly-laminar and uniform co-flowing stream of air for simple and reproducible boundary conditions. The size of the coflow was chosen in order to minimize room air entrainment while matching the coflow velocity of the previous experiments at Sandia and DLR [8]. The stainless steel fuel tube runs through the center of the entire length of the test facility, coupled on the bottom surface by a Conax packing gland. The entire facility is attached to two Velmex translation stages that allow for movement parallel to the laser beam (radial dimension = r) and perpendicular to the beam in the vertical direction (axial dimension = x). The co-flowing air is controlled using an inline flow meter mounted after the wall-mounted filters. Each fuel gas (H₂ and N₂ in this study) is supplied from compressed-gas cylinders and controlled by calibrated digital mass flow controller (Sierra Instruments, Smart-Trak 2 C). The gas streams are combined approximately 4 m prior to the main fuel tube at the base of the turbulent jet burner. An
image of the DLR H3 jet flame (50% H₂ / 50% N₂, Re = 10,000) is shown in Figure 3.16. The green lines mark the positions of data taken with the HEPBLS and discussed in Chapters 6 and 7.

Figure 3.15: Turbulent jet in-coflow-facility
Figure 3.16: Image of a DLR H3 (Re=10,000) jet flame with green dashes depicting the 10 kHz measurement locations at $x/D=10$, $20$, and $40$. 
Both the Rayleigh and Raman cameras in the imaging spectrometer are Vision Research Phantom v710 high-speed complementary metal-oxide semiconductor (CMOS) cameras shown in Figure 3.17. While scientific-grade CCD cameras have been optimized for decades to exhibit low noise, uniform pixel response and minimal nonlinearities, the widespread use of high-speed CMOS cameras within the scientific community has largely coincided with recent advances in high-speed lasers and thus is relatively new [121]. The typical application has been qualitative visualization and thus the assessment of the CMOS architecture for quantitative imaging is limited to a few studies [105], [121], [122]. In general, CMOS cameras exhibit higher levels of noise, non-uniformities, and non-linearity as compared to their CCD counterparts; however, CMOS cameras can achieve orders-of-magnitude higher framing rates as compared to CCD cameras. Each pixel of a CMOS camera sensor array has its own buffering and amplification, creating a chip where each pixel acts as an independent sensor [11]. Because of this architecture, CMOS cameras cannot be operated with on-chip hardware binning to reduce read noise like their CCD counterparts. The V710 camera sensors are constructed with an array of 1280 x 800 20-μm pixels and 12-bit pixel depth. They can be operated at 7.5 kHz at full resolution or up to 1.4 MHz at reduced 128 x 8 pixel resolution. For the 10-kHz measurements presented in this dissertation, the V710 cameras are operated with a resolution of 848 x 440 pixels.
Figure 3.17: Photographs of the Rayleigh and Raman camera setup. Both the Rayleigh (top) and Raman (middle) cameras are Phantom v710 high-speed CMOS cameras controlled by a LaVision high-speed controller (bottom). The Raman camera is coupled to a LaVision high-speed IRO, controlled by a LaVision image intensifier control unit (bottom).
The Raman camera was coupled to a high-speed image intensifier (LaVision high-speed intensified relay optics, HS-IRO). The HS-IRO used in this work consists of two stages, where the first stage contains a visible and red-enhanced sensitive photocathode (S25) and the second stage contains an S20 (blue and UV sensitive) photocathode. The S25 photocathode was chosen for its increased sensitivity (shown in Figure 3.18) in the 550-690-nm spectral range which corresponds to the vibrational Raman spectra of interest. Both stages contain a P46 “green-emissive” phosphor with primary emissions at visible wavelengths near 530 nm and a <1μs decay time. IRO systems (or image intensifiers in general) are driven by large differences in voltage. When the IRO is in the “on” position, a large negative voltage of 180V is applied to the photocathode, where incoming light is converted to electrons via the photoelectric effect. These electrons are amplified inside a microchannel (MCP) plate (0 to 900 V depending on gain setting) as they travel towards the phosphor screen, which is connected to a high voltage power supply (6 kV). The HS-IRO was controlled by an image intensifier control unit (LaVision) with a variable voltage (0-3 V) controlling the gain in the MCP and a built-in delay generator to control the photocathode voltage (creating a gate for the IRO).

As discussed above, CMOS cameras may exhibit higher levels of sensor non-linearity and non-uniformity as compared to commonly-used scientific-grade CCD cameras. In this work, it is desired to use the CMOS camera systems for quantitative measurements; hence it is important to investigate and characterize the sensor properties as shown previously within the literature see [121]. In order to determine the linearity of the Rayleigh camera and the combined v710/HS-IRO Raman camera, a series of
measurements of camera signal counts as a function of camera-incident photons were acquired. The linearity of the camera systems was determined by placing neutral density (ND) filters of known absorption [123] in front of the entrance slit to the spectrometer while keeping the incident laser energy (and therefore scattered photons) constant. While the constant laser energy ensured a constant number of scattered photons, the ND filters allowed a variation in the number of incident photons to be collected by the camera systems. Data was taken in air and non-reacting jets consisting of 42% H₂/58% air. This allows the determination of linearity curves for the integrated Rayleigh signal as well as all species-specific Raman lines except water. Data was limited to non-reacting flows as they have the least amount of signal blurring due to the increased spectral width of Raman lines at high temperatures, species signal cross-talk (Raman camera), and other interferences such as flame luminosity or fluorescence, therefore giving the most accurate measurement of signal linearity. The measured raw signals were corrected by subtracting a constant background, correcting for laser energy fluctuations, correcting for image vignetting, averaging over 272 laser pulses, and summing the binned Raman and Rayleigh signals over the entire length of the laser line. The expected linear signal ($ES_\lambda$) is determined as the product of the unfiltered measured signal ($MS_{NF}$) and the wavelength-specific transmission ($Ti(\lambda)$) of the neutral-density filters:

$$ES_\lambda = MS_{NF} \prod_{i=1}^{N} Ti(\lambda) \quad \text{Equation 3.1}$$

Results are shown in Figure 3.19. While good agreement is seen in the linear plot (top), some departure from strict linearity is observed in the oxygen measurements on the log-
scale plot (bottom), although the level of non-linearity is small. The coefficient of determination (or $R^2$) values of the measured signal as compared to the expected linear signal for each channel is shown in Table 3.1. As shown in Table 3.1, both the Rayleigh CMOS camera and the Raman CMOS/IRO camera system are linear within the experimental uncertainty of the measurement which includes shot noise and the small uncertainty in transmission properties of the individual filters.

![Figure 3.18: Red-enhanced vs. blue-enhanced photocathode quantum efficiency](image)

<table>
<thead>
<tr>
<th>Channel</th>
<th>$R^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rayleigh</td>
<td>.9985</td>
</tr>
<tr>
<td>Nitrogen Raman</td>
<td>.9970</td>
</tr>
<tr>
<td>Oxygen Raman</td>
<td>.9951</td>
</tr>
<tr>
<td>Hydrogen Raman</td>
<td>.9971</td>
</tr>
</tbody>
</table>

Table 3.1: Coefficients of determination ($R^2$) of linear fit to measured camera signal response.
Figure 3.19: Camera linearity study. Measured signal on each channel is compared to an expected linear signal as determined by Equation 3.1. The top figure is plotted on a linear scale while the bottom figure is plotted on a log scale.

Another potential issue when using high-speed imaging equipment is intensifier charge depletion [121]. Charge depletion, or a reduction in signal over a series of images for the same incident light and gain settings, occurs when the HS-IRO does not have sufficient recovery time between frames to fully charge the microchannel plate and phosphor.
screen. In Weber et al [121], a strong dependence was noted on the specific IRO, signal counts, repetition rate, and illuminated area of the photocathode; that is, charge depletion appears to be device and operating condition specific. To investigate potential charge depletion effects in the current system, the CMOS/HS-IRO combination was tested at varying repetition rates, number of incident photons (signal counts), and illuminated area of the photocathode. For the fully-illuminated and partially-illuminated (20% area) cases, the camera was illuminated with an Ulbricht sphere (LMT Luminance Standard LN 3) which was 350 mm and 750 mm away from CMOS/HS-IRO camera system, respectively. For further reduction in photocathode area illumination, a 12% illumination case was achieved by blocking the center of the Ulbricht sphere’s diffusing screen in combination with a 750-mm separation between the CMOS/HS-IRO camera system and Ulbricht sphere. For all illumination cases, an area corresponding to 51x51 pixels on the CMOS sensor was selected and averaged over ten sets of images per setting. The results were then normalized by the mean of the ten-set average signal from images 4:6. All images were taken at a voltage gain of 76 and repetition rate of 20 kHz (gain = 73 and a 10-kHz repetition were the final operating settings reported in Chapters 5-7). The results of the charge depletion study are shown in Figure 3.20, which shows that charge depletion is a major issue in the fully-illuminated camera case for this particular HS-IRO. When the average signal on images 4:6 is 2900 counts (out of a maximum of 4064), the camera registers decreasing amounts of signal every image until the intensifier can no longer charge itself after somewhere between 50 and 100 images. When the intensifier reaches a point where it can no longer keep resetting the voltage, a completely saturated

57
image is produced and all subsequent images are at or near the background signal on the CMOS camera alone. The effect of charge depletion on system response is seen in the spikes on the 2900 (blue) and 1156 (red) signal count curves. These spikes are not equivalent with what would be an average charge of 4096 because the saturated images do not appear on the same image in all 10 of the 250-image sets. With these settings (gain 76 and 20 kHz), it is seen that even with a low signal count of 174 counts on the fully-illuminated sensor, there is a 10% signal drop between image 50 and image 225 due to charge depletion.

The effects of charge depletion on a partially illuminated photocathode, as which would exist in a 1D Raman scattering imaging, are far less significant than those on the fully-illuminated sensor. Shown in the middle image of Figure 3.20 are the results from the same study repeated on a photocathode with approximately 20% of the area illuminated. While a small charge depletion pattern is seen in the 3142-count curve, both the 1362- and 314-count curves show little-to-no evidence of charge depletion. Shown in the bottom image of Figure 3.20 is a 12% illuminated photocathode, which is an estimate of the maximum total integrated signal possible in the 1D Raman scattering camera system. For this case, only a 4% difference in signal occurs between images 20:190 for a high-signal case. Due to the higher signal, gain setting, and repetition rate, this difference is expected to be an upper bound for effects of charge depletion on the results presented in this dissertation. It is further noted, that the current “red enhanced” IRO system exhibited the most significant depletion effects of any CMOS/HS-IRO systems in the TCRL.
Figure 3.20: Intensifier charge depletion study. Shown are the normalized signals for a 51 x 51 pixel block plotted versus image number for a fully illuminated photocathode (top), 20% photocathode illumination (middle), and 12% photocathode illumination on the HS-IRO/CMOS camera taken at a gain of 76 and 20-kHz repetition rate. The normalized signal count is given in the legend.
3.6 Spatial Resolution and Sampling Rates

The governing length and time scales of turbulent flows was discussed in detail in Chapter 2. Spatial resolution requirements for scalar measurements are usually given as the Batchelor scale which was defined in Equation 2.8 purely on the basis of dimensional reasoning. For temporally-correlated measurements, the highest frequency present within the flow likely corresponds to a “convective Batchelor frequency”, which is related to the Batchelor scale ($\lambda_B$) through the relation:

$$f_B = \frac{U}{2\pi \lambda_B} = \frac{U}{2\pi c} \text{Re}_L^{-3/4} \text{Sc}^{-1/2}$$

Equation 3.2

where $U$ is the local mean velocity and $c$ is a dimensional constant. Nyquist-Shannon sampling theory requires a sampling rate of more than twice the frequency being measured. Therefore, in order to resolve the smallest scalar fluctuations within the flames, the repetition-rate of the measurement needs to be at twice the characteristic frequency in the probe volume (see Table 3.2).

Spatial resolution requirements have been assessed recently by Mi and Nathan [124] in turbulent heated non-reacting jets, by Kaiser and Frank [125] in “cold” non-reacting turbulent jets and turbulent nonpremixed flames and by Wang and Clemens [37] in turbulent nonpremixed jet flames. All studies concluded that spatial resolution requirements for scalar and scalar variance were much less stringent than that for the corresponding scalar dissipation rate measurements. For example Mi and Nathan [124] concluded that spatial resolution requirements of $11\lambda_B$ were sufficient for accurately resolving the scalar and scalar variance fields. Kaiser and Frank [125] concluded that a
spatial resolution of $N\lambda_B$, where $N = 5$ to 20 depending on spatial position, was sufficient to resolve 98% of the dissipation energy. Wang and Clemens [126] used low-noise, high-resolution, temperature measurements to conclude resolution measurements of $9\lambda_B$ were sufficient to resolve 90% of the average dissipation energy. Sampling requirements also were assessed by Wang and Clemens [126], where temporal resolution requirements of $0.17f_B$ and $0.7f_B$ are required to resolve 90% of the variance and dissipation energy, respectively.

In the current work, the limiting measurement spatial resolution is defined by the laser beam thickness in the probe volume (140 $\mu$m FWHM, 280 $\mu$m $1/e^2$). In terms of temporal resolution, the HEPBLS can operate over a wide range of repetition rates, but the energy output is maximized at 10 kHz, making it a suitable choice for the high-speed Raman measurements. With these considerations, measurement locations within the turbulent flames should be chosen such that the experimental resolution and sampling rate satisfies the spatial and temporal resolution requirements identified above. For turbulent flames, $\lambda_B$ will vary considerably as a function of spatial position due to the temperature-dependent nature of kinematic viscosity. However, estimates of $\lambda_B$ and $f_B$ can be made on centerline of the turbulent nonpremixed jet flame using existing scaling relations [127]. The estimated values on centerline are expected to produce the smallest values of $\lambda_B$ and correspondingly, the highest values of $f_B$ and thus, represent the most stringent criteria for the measurements. Under self-similar conditions, a turbulent jet with

---

8 Previous work by Renfro et al. [31] measured integral timescales of OH in the DLR H3 flame along the jet centerline, where values ranged from ~0.3 ms at $x/D=20$ to ~1 ms at $x/D=40$, suggesting that a repetition rate greater than 7 kHz is required to satisfy Nyquist criteria.
heat release is described by mean flow width (δ) and centerline velocity (uc) as a function of axial position, x as:

\[ \delta(x) = 0.44x \quad \text{Equation 3.3} \]

\[ \frac{u_c(x)}{u_0} = 6.5 \left( \frac{x}{d^+} \right)^{-1} \quad \text{Equation 3.4} \]

Where \( u_0 \) is the jet-exit velocity and \( d^+ \) is the far-field equivalent momentum source diameter. For a jet issuing with uniform density and velocity from a circular nozzle with diameter \( d_0 \), \( d^+ \) is given in terms of density by:

\[ d^+ = \begin{cases} \frac{\rho_0}{(\rho_\infty)_{\text{eff}}}^{1/2} d_0, & X \leq X_s \\ \left( \frac{\rho_0}{\rho_\infty} \right)^{1/2} d_0, & X > X_s \end{cases} \quad \text{Equation 3.5} \]

where \( X \) is the mole fraction of air, where air (fluid \( \infty \)) and fuel (fluid 0) are both treated as individual fluids and \( X_s \) is its stoichiometric value. Within the flame (at mole fractions less than stoichiometric), the momentum source diameter is dependent upon the density of the fuel and an effective far-field density is defined as:

\[ (\rho_\infty)_{\text{eff}} = \rho_\infty \left( \frac{T_\infty}{(T_\infty)_{\text{eff}}} \right) \quad \text{Equation 3.6} \]

where the effective far-field temperature \( (T_\infty)_{\text{eff}} \) is calculated from extrapolating the line created by \( T_0 \) (fuel temperature) and \( T_s \) (flame temperature at stoichiometric) in mole fraction space.

\[ (T_\infty)_{\text{eff}} = T_0 + \frac{T_s - T_0}{X_s} \quad \text{Equation 3.7} \]

Beyond the flame tip (at mole fractions above the stoichiometric value), \( d^+ \) is dependent upon an effective flame density which is written as:
\[(\rho_0)_{\text{eff}} = \rho_0 \left( \frac{T_0}{(T_0)_{\text{eff}}} \right) \quad \text{Equation 3.8}\]

\[(T_0)_{\text{eff}} = T_\infty + \frac{T_S - T_\infty}{1 - x_S} \quad \text{Equation 3.9}\]

The local Reynolds number based on the mean flow width of the jet and the centerline velocity is given by:

\[\text{Re}_L = \text{Re}_\delta = \frac{\delta u_c}{v} \quad \text{Equation 3.10}\]

where the kinematic viscosity can be approximated from the mean species data available in the TNF Workshop using the Sutherland Law from [128]. The above equations can be combined to estimate the Batchelor length and time scales and frequencies on centerline as a function of axial position, as shown in Table 3.2.

The calculated Batchelor length scales and frequencies shown in Table 3.2 are calculated with Sc~1 (air) and Sc~.35 (hydrogen), which are the expected bounds under the current hydrogen-flame conditions. As shown in Table 3.2, at x/D=20, the centerline Batchelor length scale is expected to be between 140 to 240 \(\mu\)m and the Batchelor frequency should be between 18 to 31 kHz. Thus, the current measurement resolution and sampling frequencies correspond to < 2 \(\lambda_B\) and > 0.3fB, which satisfy the spatial resolution and sampling frequency requirements described above for sufficiently measuring the scalar fluctuations in both space and time. Furthermore, it is noted that for measurements at increasing radial positions, the spatial scales increase and the required sampling frequencies decrease (due to lower velocities and higher temperatures). At an axial position of x/D = 40, spatial resolution and sampling frequency requirements are satisfied at all spatial positions.
<table>
<thead>
<tr>
<th>x/D</th>
<th>T (K)</th>
<th>d’/D</th>
<th>U_/U₀</th>
<th>δ/D</th>
<th>ν_mix</th>
<th>Reδ</th>
<th>λ₁ (mm)</th>
<th>f₁ (kHz)</th>
<th>f₂ (kHz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>310</td>
<td>2.48</td>
<td>3.22</td>
<td>2.2</td>
<td>2.28E-05</td>
<td>8.42E+04</td>
<td>0.01</td>
<td>2261.75</td>
<td>1338.07</td>
</tr>
<tr>
<td>10</td>
<td>469</td>
<td>2.48</td>
<td>1.61</td>
<td>4.4</td>
<td>4.53E-05</td>
<td>4.25E+04</td>
<td>0.03</td>
<td>338.41</td>
<td>200.20</td>
</tr>
<tr>
<td>15</td>
<td>740</td>
<td>2.48</td>
<td>1.07</td>
<td>6.6</td>
<td>9.35E-05</td>
<td>2.06E+04</td>
<td>0.07</td>
<td>87.29</td>
<td>51.64</td>
</tr>
<tr>
<td>20</td>
<td>1092</td>
<td>2.48</td>
<td>0.81</td>
<td>8.8</td>
<td>1.71E-04</td>
<td>1.12E+04</td>
<td>0.14</td>
<td>31.16</td>
<td>18.43</td>
</tr>
<tr>
<td>25</td>
<td>1332</td>
<td>2.48</td>
<td>0.64</td>
<td>11</td>
<td>2.33E-04</td>
<td>8.25E+03</td>
<td>0.23</td>
<td>15.84</td>
<td>9.37</td>
</tr>
<tr>
<td>30</td>
<td>1548</td>
<td>2.48</td>
<td>0.54</td>
<td>13.2</td>
<td>3.02E-04</td>
<td>6.37E+03</td>
<td>0.33</td>
<td>9.06</td>
<td>5.36</td>
</tr>
<tr>
<td>35</td>
<td>1755</td>
<td>2.48</td>
<td>0.46</td>
<td>15.4</td>
<td>3.56E-04</td>
<td>5.39E+03</td>
<td>0.44</td>
<td>5.88</td>
<td>3.48</td>
</tr>
<tr>
<td>40</td>
<td>1655</td>
<td>0.19</td>
<td>0.03</td>
<td>17.6</td>
<td>3.12E-04</td>
<td>4.80E+02</td>
<td>3.06</td>
<td>0.06</td>
<td>0.03</td>
</tr>
<tr>
<td>45</td>
<td>1595</td>
<td>0.19</td>
<td>0.03</td>
<td>19.8</td>
<td>2.88E-04</td>
<td>5.19E+02</td>
<td>3.24</td>
<td>0.05</td>
<td>0.02</td>
</tr>
</tbody>
</table>

Table 3.2: Calculated Batchelor length scales and frequencies in the turbulent 50% N₂/50% H₂ jet flame issuing into air at Re=10,000. Jet diameter, centerline velocity, and mean flow width were calculated with scaling laws found in [127]. The mean temperature and species values were taken from flame data available through the TNF Database [9], [24]. The kinematic viscosity was calculated using the Sutherland Law found in [128]. The superscripts ¹ and ² assume a Schmidt number of 1 (air) and .35 (hydrogen), respectively.
Chapter 4: Data Processing

Much of the complexity of a Raman/Rayleigh scattering system comes in the form of data processing. This chapter will discuss the various steps involved in converting collected signal to quantitative measurements of species mole fractions, temperature, and mixture fraction in turbulent flames. The main subtopics of focus are (i) data preprocessing, (ii) the “hybrid” method of Raman/Rayleigh data analysis, and (iii) system calibration in well-characterized, near-adiabatic flames. Preprocessing includes mapping the Raman and Rayleigh cameras onto a common (and coincident) physical domain, mapping of the Raman camera pixels to the spectral domain, spatial binning, creation of “Raman channels” through spectral binning, verification of “expected” relative Raman channel-specific signal ratios under known conditions, and inspection and removal of data sets with anomalous problems such as excessive dust or beam steering.

The pre-processing is followed by the Raman/Rayleigh data analysis using the “hybrid” method as first presented in Fuest et al. [129] and described in detail throughout this chapter. Data analysis consists of (i) data preparation (i.e., background removal, corrections for pixel-to-pixel signal bleeding, and energy normalization) and (ii) Raman response matrix inversion for determining species-specific number density (or concentration). The hybrid method consists of first fitting the spectral output of the Raman camera to the theoretical Raman spectra predicted by the RAMSES code [130] in
order to create a lookup table of expected Raman spectral profiles and magnitudes at varying temperatures. This table, in addition to experimentally-determined calibration curves and a set of system-derived calibration coefficients, are used to create a library of the temperature-, species- and spatial-dependencies of each Raman channel. Through an iterative method, which is described in detail below, the inversion of the Raman-response matrix allows the determination of species concentrations and temperature as a function of spatial position. Finally, calibration constants used within the matrix inversion process are determined from ambient-temperature, non-reacting jets of air, N₂, O₂, and H₂ and stable, laminar, near-adiabatic H₂/air flames.

4.1 Preprocessing

4.1.1 Camera Mapping

Because Raman and Rayleigh data is processed concurrently and inter-dependently, it is necessary to ensure that the two data sets are correctly mapped onto a common physical space. In order to perform the mapping, a spatial mask (shown in Figure 4.1) was placed at the focal volume of the combined Raman/Rayleigh imaging system and illuminated by a neon pencil lamp (Newport 6032) and a 532-nm CW Nd:YAG laser. The spatial mask, consisting of 0.25 mm hole spacing (0.5 mm spacing around the center hole), was created via laser drilling using the focal point of a 532-nm Nd:YAG laser beam onto a strip of aluminum shim stock. The shim stock was attached to an aluminum washer and held in place by a 1” fixed lens mount. The hole diameters were measured using a microscope and were found to be approximately 50-75 µm.
Figure 4.1: Spatial mask used for mapping Raman and Rayleigh cameras. The mask was illuminated by a neon pencil lamp (for red wavelengths) and a CW Nd:YAG laser (for 532 nm). Images of the mask were used to map the Rayleigh and Raman cameras to a common physical space and to map the physical dimensions of the Raman camera to a spectral dimension.

Average results from 300 individual images acquired with both the Raman and Rayleigh cameras are shown in Figure 4.2. Light sources of two wavelength regions (i.e., “green” for Rayleigh and “red” for Raman) are necessary in order to acquire signals on both the Raman and Rayleigh cameras without removing any optics from the spectrometer. Due to the smaller physical size of the laser output as compared to the neon lamp, multiple sets of calibration images are necessary to image the entire length of the Rayleigh line segment. The Rayleigh image shown in Figure 4.2 is a summation of multiple sets of calibration images in order to display the entire Rayleigh line length. The Raman and Rayleigh cameras were mapped onto a common physical space in the spatial direction by determining the optical magnification of each camera from the images of the
mask (Figure 4.2). The imaged hole pattern also was used to determine the usable 1D segment length in the spatial direction. The center of the usable region (in the spatial direction) on both cameras was matched using the center of the closest hole visible on both cameras.

Figure 4.2: Spatial mask images in the Raman (top) and Rayleigh (bottom) cameras. These images show example results when using the spatial mask to map the cameras in physical space. Note that because the Rayleigh camera (bottom) was illuminated using a laser beam, multiple sets of images were used, with the laser hitting different portions of the mask, in order to map the entire length of the spatial dimension.
In principle, the illumination for the Raman camera could have been performed in a similar manner as the Rayleigh camera; that is, using a “red” laser output (i.e., 632-nm output of a HeNe laser). However, a neon lamp was used in order to have a set of known spectral features dispersed on the Raman camera as shown in the top portion of Figure 4.2. These individual lines allow for converting the camera pixel positions normal to the spatial dimension to spectral positions. From known neon-lamp spectral output (Figure 4.3) and known Raman shifts from H$_2$, O$_2$, N$_2$, and H$_2$O, the Raman camera pixels were mapped to a corresponding wavelength (shown in Table 4.1). A quadratic fit was used to find the specific relationship between camera pixel number and wavelength (in nm). Furthermore, the derived pixel-wavelength relationship was used to match previously-determined spectral bins from measurements at Sandia National Laboratories (shown in Table 4.2); the importance of which will be discussed below. The relationship also was used within the RAMSES code in order to give the spectral predictions associated pixel values for direct comparison with measured results.
Figure 4.3: Typical spectral output of Newport 6032 neon lamp taken from Newport Technical Reference sheets. Note that the relative intensities vary with operating conditions.
<table>
<thead>
<tr>
<th>Spectral Line</th>
<th>Pixel Number</th>
<th>Spectral Feature</th>
</tr>
</thead>
<tbody>
<tr>
<td>564</td>
<td>24</td>
<td>Hydrogen (Rotational)</td>
</tr>
<tr>
<td>580.15</td>
<td>118</td>
<td>Oxygen</td>
</tr>
<tr>
<td>585.24</td>
<td>150</td>
<td>Neon Lamp</td>
</tr>
<tr>
<td>588.18</td>
<td>166</td>
<td>Neon Lamp</td>
</tr>
<tr>
<td>594.48</td>
<td>202</td>
<td>Neon Lamp</td>
</tr>
<tr>
<td>597.55</td>
<td>220</td>
<td>Neon Lamp</td>
</tr>
<tr>
<td>602.99</td>
<td>251</td>
<td>Neon Lamp</td>
</tr>
<tr>
<td>607.43</td>
<td>277</td>
<td>Neon Lamp</td>
</tr>
<tr>
<td>607.45</td>
<td>274</td>
<td>Nitrogen</td>
</tr>
<tr>
<td>609.61</td>
<td>289</td>
<td>Neon Lamp</td>
</tr>
<tr>
<td>614.3</td>
<td>316</td>
<td>Neon Lamp</td>
</tr>
<tr>
<td>616.35</td>
<td>328</td>
<td>Neon Lamp</td>
</tr>
<tr>
<td>621.72</td>
<td>358</td>
<td>Neon Lamp</td>
</tr>
<tr>
<td>626.64</td>
<td>387</td>
<td>Neon Lamp</td>
</tr>
<tr>
<td>630.47</td>
<td>409</td>
<td>Neon Lamp</td>
</tr>
<tr>
<td>633.44</td>
<td>427</td>
<td>Neon Lamp</td>
</tr>
<tr>
<td>638.29</td>
<td>454</td>
<td>Neon Lamp</td>
</tr>
<tr>
<td>640.22</td>
<td>466</td>
<td>Neon Lamp</td>
</tr>
<tr>
<td>650.65</td>
<td>527</td>
<td>Neon Lamp</td>
</tr>
<tr>
<td>653.28</td>
<td>543</td>
<td>Neon Lamp</td>
</tr>
<tr>
<td>659.89</td>
<td>582</td>
<td>Neon Lamp</td>
</tr>
<tr>
<td>661</td>
<td>584</td>
<td>Water</td>
</tr>
<tr>
<td>667.82</td>
<td>629</td>
<td>Neon Lamp</td>
</tr>
<tr>
<td>671.7</td>
<td>652</td>
<td>Neon Lamp</td>
</tr>
<tr>
<td>683</td>
<td>723</td>
<td>Hydrogen</td>
</tr>
<tr>
<td>692.94</td>
<td>780</td>
<td>Neon Lamp</td>
</tr>
<tr>
<td>703.24</td>
<td>843</td>
<td>Neon Lamp</td>
</tr>
</tbody>
</table>

Table 4.1: Wavelengths and corresponding pixel numbers used to map the Raman-camera pixels to spectral positions.
Table 4.2: Spectral binning used on the Raman camera to match conditions at Sandia National Laboratories. The asterisks denote where the OSU binning does not match the Sandia binning wavelength due to differing fields of view on the Raman camera between the two sets of experiments

<table>
<thead>
<tr>
<th>Channel Name</th>
<th>Beginning Wavelength</th>
<th>Ending Wavelength</th>
<th>Beginning Pixel</th>
<th>Ending Pixel</th>
</tr>
</thead>
<tbody>
<tr>
<td>B3</td>
<td>689.2</td>
<td>698.6</td>
<td>758</td>
<td>848*</td>
</tr>
<tr>
<td>H₂</td>
<td>678.5</td>
<td>686.2</td>
<td>694</td>
<td>740</td>
</tr>
<tr>
<td>B2</td>
<td>666.0</td>
<td>675.2</td>
<td>620</td>
<td>674</td>
</tr>
<tr>
<td>H₂O</td>
<td>650.7</td>
<td>663.0</td>
<td>529</td>
<td>602</td>
</tr>
<tr>
<td>CH₄ (3)</td>
<td>631.8</td>
<td>643.6</td>
<td>418</td>
<td>487</td>
</tr>
<tr>
<td>CH₄ (2)</td>
<td>629.5</td>
<td>631.7</td>
<td>405</td>
<td>418</td>
</tr>
<tr>
<td>CH₄ (1)</td>
<td>624.8</td>
<td>629.4</td>
<td>377</td>
<td>404</td>
</tr>
<tr>
<td>N₂</td>
<td>601.9</td>
<td>610.2</td>
<td>245</td>
<td>293</td>
</tr>
<tr>
<td>CO</td>
<td>595.1</td>
<td>601.8</td>
<td>206</td>
<td>244</td>
</tr>
<tr>
<td>B1</td>
<td>585.2</td>
<td>592.6</td>
<td>149</td>
<td>191</td>
</tr>
<tr>
<td>O₂</td>
<td>576.3</td>
<td>582.3</td>
<td>99</td>
<td>133</td>
</tr>
<tr>
<td>CO₂ (2)</td>
<td>571.2</td>
<td>576.2</td>
<td>70</td>
<td>98</td>
</tr>
<tr>
<td>CO₂ (1)</td>
<td>564.2</td>
<td>571.0</td>
<td>30</td>
<td>69</td>
</tr>
<tr>
<td>C₂</td>
<td>552.2</td>
<td>563.1</td>
<td>1*</td>
<td>24</td>
</tr>
</tbody>
</table>

4.1.2 Spectral Fitting of Raman Images with RAMSES

In order to use the “hybrid” method of data reduction as described in Fuest et al. [129], a spectral library composed of temperature-dependent Raman channels must be created. RAMSES, a software for the calculation of Raman spectra provided by TU Darmstadt [130], was used to produce the spectral library. Matching the output of the library with the spectral features collected on the Raman camera requires both the wavelength-to-pixel number conversion (described above) and an “instrument function” in order to assign a line shape to the spectral features predicted in the program that can be directly
compared to the measured, spectrally-distributed camera signals. While the RAMSES code can predict the species-specific spectral distribution due to local temperature and pressure, the measured distribution on the Raman camera in the spectral dimension is a function of the species-specific spectral distribution and the instrument function. The “instrument function” represents the influence of the measurement system on the perceived spectral distribution of the Raman species due to the intensity distribution of the laser beam and the finite pixel (spectral) resolution of the detection system.

The common choice of instrument function is the measured intensity profile from the Rayleigh camera. When using the same camera type for both Raman and Rayleigh data collection, the Rayleigh camera gives the convolution of the intensity distribution of the laser beam and the camera response function, which is an appropriate approximation of the instrument function of the Raman camera in the spectral dimension without the consideration of the wavelength-dependent Raman shifts. The reason that the measured Rayleigh scattering intensity distribution can be used as the expected instrument function (in the spectral dimension) arises from the fact that the spectral resolution of the Raman camera is low. First, consider the case of a monochromatic excitation beam at an infinitesimal focus; that is, there is no intensity distribution in physical space. For this case, the measured distribution of the Raman signal in the spectral dimension would be equivalent to the Raman-scattered spectral lineshape, which can be calculated directly by RAMSES. Next consider the case of an excitation beam with a finite spectral distribution (or linewidth) at an infinitesimal focus. For this case, the measured distribution of the Raman signal in the spectral dimension would be governed by the convolution of the
laser spectral lineshape and the Raman-scattered lineshape. Next consider the case of monochromatic laser light (no spectral lineshape) with a finite intensity distribution (in physical space) interacting with the Raman-active species. For this case the measured intensity distribution on the Raman camera in the spectral dimension would be governed by the convolution of the spatially-varying laser intensity distribution and the Raman-scattered spectral lineshape since the spectral dispersion from the grating is relative to the incident point on the grating, which would vary as the intensity distribution in physical space.

For the two laser excitation sources considered, the laser spectral linewidth is approximately 1 cm$^{-1}$ and 0.1 cm$^{-1}$ for the 10-Hz, Q-switched Nd:YAG and the HEPBLS, respectively. These linewidths should be compared with the Raman camera spectral resolution which is determined from the camera pixel-to-wavelength relationship displayed in Table 4.1. From this data, the Raman camera spectral resolution is $\sim 0.17$ nm/pixel or $\sim 5$ cm$^{-1}$/pixel. This implies that the spectral distribution of the excitation laser is much less than one pixel in the spectral dimension on the Raman camera, while for the current 1:1 imaging, the nominal 200 $\mu$m ($1/e^2$) laser beam thickness (or width of the intensity distribution) comprises approximately 10 pixels in the spectral dimension (once dispersed by the grating). For the current experimental setup, the convolution of the laser intensity distribution and camera response function is the dominant source of the instrument function.

Though an intensifier was used in addition to the Raman camera, the measured Rayleigh scattering intensity distribution is still considered the best instrument function.
available and was used to create the spectral library. This was chosen due to the superior spatial resolution and signal-to-noise of the Rayleigh camera. Direct measurements from the Raman camera (i.e., N\textsubscript{2} Raman profile) were not used because of signal bleeding from the intensifier (discussed below). Results from a fit to N\textsubscript{2} and O\textsubscript{2} room-temperature air using the Rayleigh camera-derived instrument function are shown in Figure 4.4. The measured spectrum is a single pixel (in the spatial dimension) slice of the resulting Raman signal averaged over 270 10-Hz laser shots. For this particular example, the predicted spectrum for each species was scaled by matching the peak values of the predicted spectra to corresponding peaks on the measured Raman spectrum. It should be noted that when processing single-shot flame data, the predicted spectra are not arbitrarily scaled to match the experimental data. One set of species-specific calibration constants are determined experimentally and are used with the measured Raman signals and predicted Raman spectra from RAMSES to determine individual species concentrations. The simple scaling to match peaks between measured and simulated spectra was done here for the purposes of comparing the predicted and measured spectral locations.

While Figure 4.4 shows that there is good agreement in the spectral locations between the predicted and measured spectrum when using the pixel-wavelength relationship described in Section 4.1, there are larger wings (wider spectral bandwidth) on the spectral features of the measured data as compared to that predicted by RAMSES. This effect is attributed to signal gradient blurring which is known to exist when using two-stage high-speed image intensifiers [99]. In previous high-speed imaging studies,
Gordon et al. [99] found that in the regions of steep gradients, there was apparent signal diffusion or high signal values “bleeding” onto adjacent lower-signal pixels, which manifest themselves as gradient spreading, both as a local modular transfer function (MTF) effect and a non-local blurring. In their work, the effects were most noticeable at the lower (relative) signal levels, which is consistent with that observed in Figure 4.4 in the “wings” of the spectral profiles.

![Air at 300K](image)

Figure 4.4: Results from a RAMSES fit to measured Raman spectra in air at 300 K using the Rayleigh signal in room-temperature air as the instrument function. The calculated spectrum (red) was fit to the averaged measured spectral profile (blue) by matching the peak signal on all species channels.

Differences in the instrument function between the unintensified Rayleigh camera and the intensified Raman camera are expected due to the signal gradient blurring;
however, an attempt was made to reduce the effects of the gradient blurring and the error between the measured and simulated spectra. From examining the spectral fits under various operating conditions, it was hypothesized that the increased signal in the spectral wings has a linear dependence on the peak signal of nearby spectral features. This would imply that the broadening of the measured spectral profile is dependent on the local signal conditions (which will fluctuate) and furthermore that the broadening of any given species-specific profile is a function of all nearby species profiles. For example, this would imply that the wings of the oxygen spectral feature are dependent not only on the peak of the oxygen signal, but also the peak of the nitrogen signal.

In order to create an empirical correction for the signal “bleeding” on the intensified Raman camera, Raman scattering from a single species, nitrogen, was imaged. Neutral density filters (Thorlabs) were placed in front of the entrance slit to the spectrometer in order to achieve various signal strengths on the Raman camera. The expected nitrogen line shape (calculated using RAMSES and scaled to the peak measured signal) was subtracted from the averaged measured signal and normalized. The results are shown in Figure 4.5 (left). Despite the order-of-magnitude difference in peak signal, the resulting shape of the error (which corresponds to the difference in signal in the wings of the spectral shape) is consistent for all incident signal level cases considered. These results suggest that the increased signal within the wings of the spectral profiles is nearly linear with peak signal strength.
Figure 4.5: A comparison of the measured and predicted Raman signal of nitrogen for various incident signals. The predicted RAMSES profile for nitrogen was subtracted from the measured N$_2$ Raman profile (left). An empirical correction in the shape of a triangle 320 pixels long with a peak magnitude of 2% of the peak N$_2$ Raman signal also was applied (right).

While various corrections for the signal bleeding were considered, any correction would need to be robust and applied to every single-shot image prior to processing, making a simple correction preferred to more complicated options. A triangular correction was used due to its simplicity and efficacy in reducing the error between the measured and predicted spectra. A triangular correction 320 pixels in length with a peak value corresponding to 2% of the peak Raman signal strength, as determined by the normalized signal near the feature center in Figure 4.5 (left), was subtracted from the nitrogen Raman signal. Figure 4.5 (right) shows the calculated error between the corrected normalized nitrogen signal and the calculated RAMSES spectra. A notable reduction in the difference between the measured and calculated N$_2$ Raman spectrum is shown.
When applying the empirical correction to an image with more than one species present, a triangular correction was calculated for each species-specific spectral feature. The “individual” triangular corrections were added together to apply a total “signal bleeding” correction. Figure 4.6 shows the same profiles in air as previously shown in Figure 4.4 as well as the measured profile with the “bleeding” correction applied. While the corrected measured signal (pink) still exhibits a broadening of the spectral profile as compared to the predicted spectrum (red), improvements over the “uncorrected” measured signal are shown. While applied to all flows, the “bleeding” correction is most significant in cold flows because of the high signal levels. For single-shot images, the peak signal on each species channel was calculated before pixel binning (discussed below), allowing for the correction to be calculated after the data was software binned. This approach, rather than preprocessing of the correction, allowed for the correction to be modified if necessary during calibration if deemed necessary. While this approach was somewhat successful in reducing the baseline signal in cold flows where signal counts were high, it was less effective in low-signal environments.
Figure 4.6: Results from RAMSES fit to measure Raman spectra in air at 300 K. The calculated spectrum (red) was fit to the averaged measured spectral profile (blue) by matching the peak signal on all species channels. An empirical correction to account for the “pixel bleeding” on the intensifier was subtracted from the average measured profile (pink).

4.1.3 Spectral and Spatial Binning

In order to process the Raman signal, each Raman image must be broken into distinct “Raman channels” (via pixel binning) corresponding to the signal expected from each individual species. The signal in each channel is summed over a select number of pixels in the spectral direction to give a total signal for each species channel for a single pixel in the spatial dimension. The number of pixels in each channel is chosen to correspond to the wavelengths covered by the Raman channels used at the Combustion Research Facility (CRF) at Sandia National Laboratories (provided by Robert S. Barlow and shown for this experiment in Table 4.2). By matching the wavelengths covered by the channels
at Sandia, it was possible to use the “Sandia polynomials”, an experimentally-determined set of expressions (cast in polynomial form) describing the expected Raman signal from each major species and species cross-talk on each channel over a range of temperatures from 290 K to 2500 K [131]. The importance of the wavelength matching comes from the fact that the Raman signals and cross-talk (of each channel) are highly temperature dependent. The temperature dependence of each parameter (10 total in the current work) have been determined at the CRF in a detailed set of calibration experiments consisting of cold gas flows, electrically-heated flows, and well-characterized flames that have been performed over several years. However, it is noted that the CRF setup utilizes scientific-grade CCD cameras which permits “hardware” or “on-chip” binning prior to readout which significantly increases the signal-to-noise ratios of the measurements. At the CRF, the Raman camera utilizes hardware binning in both the spatial and spectral dimension, thus the “Sandia polynomials” are determined with and valid only for fixed wavelength ranges.

As shown in Table 4.2, there is at least one channel for each species. In the case of CO$_2$, the separate channels correspond to two distinct vibrational Raman lines. The multiple channels for methane are due to the complex Raman signature of the C-H stretch bond. The C$_2$ channel corresponds to a background channel often used to measure the influence of C$_2$ fluorescence on all measurement channels in the case of hydrocarbon flames. In this work, only hydrogen flames are considered, thus only the channels corresponding to oxygen, nitrogen, water, hydrogen, and background signals were used. However, by keeping the same wavelength ranges for each Raman channel as used at the
CRF, the current data analysis can be extended to hydrocarbon flames and the measurement of CH$_4$, CO$_2$, and CO.

In order to achieve higher signal-to-noise ratios, the acquired signals on both the Raman and Rayleigh channels were software binned in the spatial direction as well, yielding “superpixels”, which for the Raman camera denotes the effective binning in both the spatial and spectral dimensions. While increased binning in the spatial direction will yield higher signal-to-noise ratios (SNR), the increased SNR is balanced with reduced spatial resolution. It is noted that CMOS camera architecture does not permit hardware binning, thus all pixel binning in reference to the OSU high-speed system refers to software binning; that is pixel binning of the final digitized signals after readout. To examine the effects of the software binning (in the spatial dimension), data taken at 10 Hz was processed with 80 µm (4 pixel binning), 160 µm (8 pixel binning), and 320 µm (16 pixel binning) resolution on the Raman camera and 80 µm binning on the Rayleigh Camera. Figure 4.7 plots the SNR (after all data processing) for the nitrogen channel in both air and stoichiometric flame conditions. The SNR is defined as the mean value from 272 individual images divided by the standard deviation of the 272 images. Another test was conducted where the effects of binning the Rayleigh camera on the Raman and Rayleigh SNR was examined since the final results are a function of both the Rayleigh and Raman signals. While there was a small increase in Rayleigh channel SNR with increased binning, there was no perceptible effect on the species mole fractions. A small Rayleigh channel binning was chosen in order to minimize errors in mapping the two cameras.
Figure 4.7 shows that the SNR increases (nonlinearly) with increased pixel binning, but at a rate lower than expected. In the reported results, software binning of 8 pixels in the spatial direction (160 µm) on the Raman camera and 2 pixels in the spatial direction (40 µm) on the Rayleigh camera was used to balance SNR with spatial resolution. This resolution also was chosen for the turbulent flame results presented in Chapter 7 in order to calculate the autocorrelation functions as the cross-correlation of two adjacent 160-µm pixels. This was done in order to decrease the effect of the electronic shot noise on the derived autocorrelation functions by using two “independent” detectors (here two “superpixels”) as discussed in [14], [132], though no verification of a decrease in noise is presented here. Images were binned in the spatial direction starting at the center of both the Raman and Rayleigh cameras (to minimize mapping error).

Figure 4.7: Signal-to-noise ratio (SNR) of nitrogen with varying spatial resolution via software binning. The Raman and Rayleigh spatial resolutions were matched.
After spatial binning, the data was comprised of three-dimensional matrices consisting of channel number (or pixel number in the case of Rayleigh signal) x spatial position x image number. A separate three-dimensional matrix containing the maximum signal in each species bin on the Raman camera prior to spectral binning was created in order to apply the signal “bleeding” correction discussed in Sec 4.1.2. Finally, it is noted that the measured signal from the CMOS camera used for laser energy measurements (Rayleigh scattering in cold air as described in Chapter 3) was summed along the entire spatial dimension to give one relative laser energy value for each image.

4.1.4 Raman Channel Signal Verification

Prior to processing the Raman/Rayleigh data with the hybrid method, mean signals from select calibration flows (see Chapter 5) were examined for expected relative signal strength on each Raman channel. Based on the species-specific Raman cross-section, the wavelength-specific quantum efficiency (QE) of the HS-IRO, and the known mole fraction of the species in question, a first-order calculation of the expected Raman signal ratios of each species was performed. These calculations serve as “sanity checks” and potential flags for errant data collection. An example of this calculation from measurements within a non-reacting flow of $0.59\text{N}_2/0.16\text{O}_2/0.25\text{H}_2$ is shown in Table 4.3.

The metric of interest is the comparison between the expected signal ratio between species $i$ and $\text{N}_2$ and the measured signal ratio. The expected signal ratio is defined as:

$$\frac{S_i}{S_{\text{N}_2}} = \frac{X_i \sigma_{\text{Ram},i} \text{QE}(\lambda)}{X_{\text{N}_2} \sigma_{\text{N}_2} \text{QE}(607 \text{ nm})} \quad \text{Equation 4.1}$$
where $X_i$ is the mole fraction of species $i$, $\sigma_{\text{Ram},i}$ is the Raman-scattering cross section of species $i$, and $\text{QE}(\lambda)$ is the wavelength-specific quantum efficiency of the HS-IRO. In the example shown in Table 4.3, the oxygen-to-nitrogen signal ratio is as expected from the known mole fraction, Raman scattering cross sections and HS-IRO quantum efficiencies. The hydrogen-to-nitrogen signal ratio is approximately 7% lower than expected; however, it is noted that these preliminary signal checks do not consider crosstalk, signal bleeding, or any other preprocessing. Again, these calculations serve as a preliminary vetting of the data and the example case shown in Table 4.3 would be deemed as valid and processed with the hybrid method as discussed below.

<table>
<thead>
<tr>
<th>Species</th>
<th>N2</th>
<th>O2</th>
<th>H2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Raman cross-section cm$^2$/sr x 10$^{-30}$</td>
<td>0.46</td>
<td>0.65</td>
<td>0.943</td>
</tr>
<tr>
<td>IRO QE (nm)</td>
<td>14% (607 nm)</td>
<td>16% (580 nm)</td>
<td>12% (683 nm)</td>
</tr>
<tr>
<td>Unlit Flame 12 at 10 Hz</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Mole Fraction</td>
<td>0.59</td>
<td>0.16</td>
<td>0.25</td>
</tr>
<tr>
<td>Background Corrected Signal</td>
<td>2.9E+04</td>
<td>1.2E+04</td>
<td>2.0E+04</td>
</tr>
<tr>
<td>Expected Signal $\frac{S_1}{S_{N2}}$</td>
<td>1</td>
<td>.43</td>
<td>.75</td>
</tr>
<tr>
<td>Measured $\frac{S_1}{S_{N2}}$</td>
<td>1</td>
<td>.43</td>
<td>.70</td>
</tr>
</tbody>
</table>

Table 4.3: Example Raman-channel signal verification for a non-reacting flow of 0.59 N$_2$ / 0.16 O$_2$ / 0.25H$_2$. 

85
In addition to verifying the expected signals on each Raman channel, high-speed (10-kHz) data acquired with the HEPBLS was examined for charge depletion effects by monitoring the collected signal as a function of “shot number” within a pulse burst. The results of raw N₂ Raman signal in air (averaged over 10 bursts) are shown in Figure 4.8. Plotted are the energy-corrected, normalized signal for both the Raman (HS-IRO + CMOS) and Rayleigh (CMOS only) cameras. Each trace is normalized by the average signal value at shot 100 in the burst. It should be noted that the acquired Raman and Rayleigh scattering signals are not constant from the beginning to the end of the burst. Figure 4.8 shows that there is a small dependency of signal on shot number in both the Raman and Rayleigh data. “Shots” < 20, were not processed because of a rapid increase in pulse energy output from the HEPBLS at the beginning of the burst. The Raman data shows the same initial trend with an additional caveat that it takes a few more shots to reach a steady-state value than the Rayleigh data. This is likely due to properties of the HS-IRO, although no evidence of charge depletion is seen due to the finite duration of the pulse burst and associated signal acquisition. For comparison, an example of the calculated temperatures (from ambient air) within a single burst based on the Raman and Rayleigh signals is shown in Figure 4.9. There is no visual evidence that the measured temperature has any dependence on shot number with the exception of the very beginning of the burst where laser energy is very low. This portion of the burst is not used in any data analysis.
Figure 4.8: Signal vs. HEPBLS shot number averaged over ten pulse bursts in air and corrected for laser energy. Data was acquired at 10 kHz.

Figure 4.9: Temperature vs. shot number of air in a single burst as calculated using the hybrid method. Data was acquired at 10 kHz.
4.2 “Hybrid” Method

Historically, there are two different approaches to processing Raman data, polynomial matrix inversion methods [37], [47], [133] and spectral fitting methods [41], [130], [134]. The polynomial matrix inversion method is an iterative method that calculates species concentrations by inverting a matrix of temperature-dependent calibration constants derived from extensive calibration curves. These calibration curves or, “Sandia polynomials”, are the result of detailed spectral measurements taken of cold gases, electrically-heated gases, and well-characterized near-adiabatic reacting flows of all expected combustion species and background fluorescence. While the matrix inversion method allows for the relatively simple processing of the concentration of species and high signal-to-noise by taking advantage of on-chip binning, the accuracy of the method is dependent upon the quality of the calibrations, which take extensive time and resources to carry out. As cross-talks and background interferences at high temperatures are determined experimentally in flames, there is large uncertainty in their values due to the presence of many species [129]. Alternatively, the spectral fitting method determines species concentrations by fitting experimentally-measured spectra to a library of theoretically-calculated, temperature-dependent expected rotational and vibrational Raman transitions. The calculated Raman transitions are convolved with an apparatus function of the detection unit (to give them their expected spectral width) and calibrated with a gas sample at known conditions. While the spectral fitting method more easily corrects for background interferences and species-crosstalk, it requires approximate spectrum for complex molecules [41] and significantly more computation time to fit the
experimental spectra for every data point [129]. As described below, the hybrid method proposed in Fuest et al. [129] combines the strengths of both the matrix inversion and spectral fitting approaches.

The hybrid method of Raman/Rayleigh data processing [129] relies upon both the matrix inversion of calibration-dependent system response [133] as well as spectral fitting to theoretically-calculated Raman spectra [130]. While this method has been used extensively by researchers at the Combustion Research Facility at Sandia National Laboratories (for examples see [77], [79], [135]–[137]), it has not previously been employed with a high-speed CMOS camera-based collection system. The extensive calibrations necessary to use the matrix inversion method were taken using a high-resolution, low-noise CCD-based experimental setup at the CRF and applied to the high-speed CMOS-based system within the Turbulence and Combustion Research Laboratory at Ohio State. Through the use of the hybrid method, only minimal calibrations of the Ohio State system were necessary to ensure accurate results.

4.2.1 Data Preparation

All data processing and preprocessing was performed using Matlab-based code, which included adaptations and expansions from both a matrix inversion code written by G. Wang [37] and a Labview-based hybrid method code used at the CRF [135], [136]. After preprocessing the raw image files into binned “superpixels” corresponding to the wavelength-specific Raman channels defined by the CCD camera-based, on-chip binning process at the CRF, the image files for an entire data set are loaded into Matlab. The data
structure contains the number of images, path and file names, any image numbers that reach saturation (suggesting that dust particles or laser-induced plasma from laser breakdown was present), a 1D array for laser energy, and the 3D matrices for Raman signal, maximum Raman signal prior to spectral binning, and Rayleigh signal.

The first step of processing is to remove a representative “darkfield” from the image set. For high-speed data taken with the HEPBLS, the darkfield image is an average of five images taken at the “beginning” of the burst prior to the laser reaching the test-section in order to reduce the effects of darkfield shift over time [105]. Only the five images at the beginning of the burst were used because images from the intensified Raman camera after the burst contain residual signal on the Raman channels, particularly in high-signal, cold flow regions. For the 10-Hz data, a separate darkfield image is acquired with the laser blocked and subtracted from the image set. The second step is to remove a “background” from the Raman images. A background for the measured Raman signals is determined from the signal present on the B3 background channel (see Table 4.2). Because no Raman-active species have Raman shifts in this spectral region, the B3 signal is a measure of the background from fluorescence, flame luminosity, or other stray light on Raman camera. Due to the proximity of B3 to the hydrogen channel, signal bleeding from the hydrogen channel onto B3 is expected. Care was taken to remove the effects of the signal bleeding from the hydrogen channel to B3 prior to using this value as a background subtraction from all other Raman channels.

The next step is a correction for “signal bleeding” within the Raman data as discussed in Section 4.1.2 with special attention given to the effects of the background
signal. Consider Figure 4.10, where the blue curve shows a theoretical signal due only to background effects that were not subtracted off when subtracting the darkfield. If signal bleeding were subtracted based only on the maximum signal on the channel without any correction, the red triangle (top) would be subtracted from the blue signal. This would give a dip in the signal due to the bleeding correction. By subtracting out a background (or base signal from the B3 channel) prior to calculating the signal bleeding correction (bottom), no correction is deemed necessary for this example. Since the flames studied in this work are hydrogen-fuel based, the carbon channels (CO₂, CO, and CH₄) were turned off in all calculations, including the calculation of “signal bleeding” from the CO₂, CO, and CH₄ channels to adjacent channels. Therefore the total signal bleeding correction was based only on the summation of the calculated “correction triangles” for H₂, H₂O, O₂, N₂, and the rotational H₂ (found in the C₂ channel). As noted in Section 4.1.2, the maximum correction due to signal bleeding was 2% of the maximum value on any given species channel and decreased linearly to zero over 175 pixels in either direction. The total value of a correction within a given Raman channel was summed based on the previously-established wavelength-specific “superpixels”. This correction was then subtracted from the darkfield- and background-corrected Raman signals.
After the “signal bleeding” correction, the Raman and Rayleigh data were corrected for laser-energy fluctuations. The Raman data is then parsed from 14 Raman channels to 9 (as shown in Table 4.4) to remove unnecessary and concatenate species-redundant channels while the Rayleigh data is summed over columns perpendicular to the spatial direction (creating a 2D matrix of spatial dimension x image number). A normalization map is then applied to both the Raman and Rayleigh data in order to correct flat field effects that arise from camera sensor and photocathode non-uniformity, signal vignetting, and bowing effects that arise from the Raman/Rayleigh light transmitting through the entrance slit of the imaging spectrometer. The flat field normalization is created from imaging uniform fields of non-reacting flows (and high-temperature water in a flat flame) for the Raman channels of interest. For the case of varying spatial resolutions, the Rayleigh camera is then mapped onto the Raman camera resolution (e.g., 40 µm Rayleigh resolution is mapped to 160 µm Raman resolution).

Figure 4.10: Sketch of potential signal bleeding subtraction from background effects.
Finally, the Raman channel matrix is updated again with the inclusion of a CO laser-induced fluorescence (LIF) channel in order to be consistent with the hybrid method as used at the CRF. In this work, no simultaneous LIF measurements are performed so the CO-LIF channel is inputted as an array of zeros. Both the Raman and Rayleigh matrices are smoothed with a 3-point, moving-average filter prior to matrix inversion.

<table>
<thead>
<tr>
<th>Original Raman Channels:</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
</tr>
<tr>
<td>C2</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Updated Raman Channels After Combining Channels</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
</tr>
<tr>
<td>CO2</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Final Raman Channels After Adding CO-LIF Channel</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
</tr>
<tr>
<td>CO2</td>
</tr>
</tbody>
</table>

Table 4.4: Raman channel progression. For the middle channels, duplicate channels were summed and background channels 1 and 2 were removed from the Raman data matrix. For the bottom (final channels), a CO-LIF channel was inserted into the matrix. For the current data, this channel consists of zeros.

4.2.2 Raman-Response Matrix Inversion

Generally speaking the relationship between the collected signal ($S_{\text{Ram}}$) and the species concentration ($c$) is given by:

$$S_{\text{Ram}} = C(T)I_{0}c$$  

Equation 4.2

where $S_{\text{Ram}} = (S_{\text{Ram,1}}, S_{\text{Ram,2}}, \ldots, S_{\text{Ram,n}})$ is a signal vector describing the collected signal on the Raman camera at a spatial position as a function of incident laser energy; $c = (c_{1},$
\( c_2, \ldots, c_n \) is a concentration vector at a given spatial position as a function of Raman channel \( i = 1 \) to \( n \), and \( C(T) \) is the \( n \times n \) “Raman response” matrix, which is a function of temperature, \( T \) and all species mole fractions [133], [138]. The signal on each Raman channel is a combination of the Raman scattering response of the channel-specific species (diagonal terms of \( C \)) and signal contributions from the cross-talk from all other species and additional sources of interference (off-diagonal terms of \( C \)). The hybrid method consists of first fitting the spectral output of the Raman camera to the theoretical Raman spectra predicted by the RAMSES code in order to create a lookup table of expected Raman spectral shapes and relative magnitudes at varying temperatures. This table, in addition to experimentally-determined calibration curves (“Sandia polynomials”) and a set of system-derived calibration coefficients are used to create a library of the temperature- and spatial-dependencies of the \( C \)-matrix elements. Using \( S \) and \( C \), species number densities are obtained through matrix inversion. These calculations are iterative in nature since \( C \) is a strong function of temperature, which is unknown and the process is closed once the Rayleigh temperature measurement converges.

An initial estimate of the temperature in the measurement volume is made based solely on the signal from the Rayleigh channel (assuming the chemical makeup of air at a pressure of 101.325 kPa). The temperature is determined from Equation 4.2, where the reference condition is air at 294 K and 101.325 kPa and is used within the \( C \)-matrix to determine individual species concentrations through matrix inversion. The Raman response matrix (\( C \), shown in Figure 4.11) begins as a 10 x 10 matrix of zeroes representing the expected response of each Raman channel and the effects of each Raman
channel (from Table 4.4) on all other Raman channels (see Equation 3.2). Based on the contents of a flag file, which lists all RAMSES active-matrix locations (see Table 4.5), the species-specific Raman scattering cross-sections or cross-talk parameters for the calculated temperature are interpolated from the RAMSES tables and multiplied by a system-specific calibration factor determined from calibration flows and flames (see Section 3.3). After the C-matrix elements based on the RAMSES calculations have been populated (Figure 4.11), the remainder of the active-matrix elements are populated using the experimentally-determined, temperature-dependent system response curves referred to as the “Sandia polynomials”. While RAMSES spectra are available for all species in hydrogen flames (not true for hydrocarbon flames), some crosstalk terms are less accurate than the empirically-determined temperature dependencies (i.e., H₂O and H₂ onto O₂). This set of polynomials predicts the signal for each element based on extensive calibrations from the CRF Raman/Rayleigh/CO-LIF system. The right most column of Table 4.5 lists whether the active-matrix element is derived from RAMSES or the Sandia polynomials. The remaining “inactive” matrix elements are left as zeroes.

The populated Raman-response matrix, the assumed temperature, and measured signals are used to estimate the concentrations of individual species in the measurement volume. The individual species concentrations (cᵢ) are summed to give a total concentration in the probe volume and the mole fraction of each individual species (Xᵢ) is estimated as \( Xᵢ = cᵢ/(\Sigma cᵢ) \). An updated mixture-averaged Rayleigh cross-section is calculated by summing the individual species mole fractions multiplied by the species-specific Rayleigh scattering cross-sections shown in Table 2.1. It should be noted that for
the hydrogen-based flames used in this work, the Rayleigh cross-sections for all carbon-containing species within the C-matrix are set to zero. Using the updated mixture-averaged Rayleigh scattering cross section, a new and more accurate estimate of the Rayleigh temperature, C-matrix, and species concentrations are calculated. The process iterates until the Rayleigh temperature (using updated species concentrations in the test volume) converges to a user-prescribed tolerance, which is set to 1K in this work.

After the Rayleigh-based temperature iterations are complete (generally 1-3 iterations), a perfect-gas temperature is calculated based on the total species concentration, referred to as the “Raman temperature”. Mole and mass fractions also are calculated from the final converged concentration data and the equivalence ratio is calculated with Equation 4.3 and the mixture fraction is calculated with Equation 4.4.

\[
\phi = \frac{0.5X_{H_2}O + 0.5X_{H_2}}{X_{O_2} + 0.5X_{H_2}O} \quad \text{Equation 4.3}
\]

\[
\xi = \frac{(Y_{H} - Y_{H,2})/2w_H + (Y_{O} - Y_{O,2})/w_O}{(Y_{H,1} - Y_{H,2})/2w_H + (Y_{O,1} - Y_{O,2})/w_O} \quad \text{Equation 4.4}
\]
\[ C(T) = \begin{pmatrix}
C_{CO_2} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & C_{O_2} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & C_{CO} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & C_{O_2} \ll N_2 & 0 & C_{N_2} & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & C_{CH_4} & 0 & 0 & 0 & 0 & 0 \\
0 & C_{O_2} \ll H_2O & 0 & C_{N_2} \ll H_2O & 0 & C_{H_2O} & C_{H_2} \ll H_2O & 0 & 0 & 0 \\
0 & C_{O_2} \ll H_2 & 0 & 0 & 0 & C_{H_2O} \ll H_2 & C_{H_2} & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & C_{CO_{LIF}} \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & C_{B_3}
\end{pmatrix} \]

Figure 4.11: 10 x 10 Raman response matrix \((C)\) used in this research.

<table>
<thead>
<tr>
<th>i</th>
<th>j</th>
<th>Ramses On/Off</th>
</tr>
</thead>
<tbody>
<tr>
<td>CO2</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>O2</td>
<td>2</td>
<td>2</td>
</tr>
<tr>
<td>CO</td>
<td>3</td>
<td>3</td>
</tr>
<tr>
<td>N2</td>
<td>4</td>
<td>4</td>
</tr>
<tr>
<td>H2O</td>
<td>6</td>
<td>6</td>
</tr>
<tr>
<td>H2</td>
<td>7</td>
<td>7</td>
</tr>
<tr>
<td>O2 \ll CO2</td>
<td>2</td>
<td>1</td>
</tr>
<tr>
<td>CO2 \ll O2</td>
<td>1</td>
<td>2</td>
</tr>
<tr>
<td>CO \ll N2</td>
<td>3</td>
<td>4</td>
</tr>
<tr>
<td>CO2 \ll H2</td>
<td>1</td>
<td>7</td>
</tr>
<tr>
<td>O2 \ll H2</td>
<td>2</td>
<td>7</td>
</tr>
</tbody>
</table>

Table 4.5: RAMSES active matrix locations. This table lists the possible locations in the C-matrix where elements can be derived from RAMSES calculations. The subscripts i and j refer to the location in the C-matrix and On/Off refers to whether the RAMSES calculation is used.
Chapter 5: Calibration with Near-Adiabatic Flames

The RAMSES library provides temperature and spatial dependencies of the Raman-response matrix, but calibrations are still needed in order to determine their absolute values. However, it is noted that the level of calibration within the “hybrid method” is significantly reduced as compared to the matrix inversion method. As noted above, the diagonal terms of the Raman-response matrix are populated by species-specific calibration constants (referred to here as $C_{\text{species}}$) multiplied by temperature-dependent Raman cross-sections determined from RAMSES output or the Sandia polynomials. The crosstalk constants multiplied by the associated Raman cross-sections populate the off-diagonal terms. The location of the calibration constants within the Raman-response matrix is shown in Figure 4.15.

In this research the necessary calibration constants ($C_{\text{species}}$) used within the C-matrix are determined from ambient-temperature, “cold”, non-reacting flows of air, $N_2$, $O_2$, and $H_2$ and stable, laminar, near-adiabatic $H_2$/air flames (Hencken burner, see Section 3.4.2) using the OSU high-speed Raman/Rayleigh imaging system. Flow conditions for the set of calibration hydrogen/air flames are shown in Table 5.1. The “cold”-flow conditions represent a subset of the flame conditions shown in Table 5.1 where the reactant gas streams are never ignited. The Gordon and McBride NASA Equilibrium Chemistry Code [139] was used to calculate the expected temperature and species
concentrations for the flow conditions described in Table 5.1. In order to predict the near-adiabatic behavior of the Hencken flames with minor heat losses, equilibrium species concentrations were determined with flame temperatures 35 K below the adiabatic flame temperature. The 35-K degree offset from adiabatic conditions was chosen based on conversations with R.S. Barlow, the results of N₂ CARS measurements in similar Hencken flames [120], and other estimates given for near-adiabatic burners [63], [131], [140].

<table>
<thead>
<tr>
<th>Name</th>
<th>Equivalence Ratio</th>
<th>Flame Temperature</th>
<th>Species Mole Fractions of Products</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Φ</td>
<td>(K)</td>
<td>(X_{O_2})</td>
</tr>
<tr>
<td>Flame 4</td>
<td>0.27</td>
<td>1083</td>
<td>1.44E-01</td>
</tr>
<tr>
<td>Flame 5</td>
<td>0.34</td>
<td>1252</td>
<td>1.29E-01</td>
</tr>
<tr>
<td>Flame 6</td>
<td>0.41</td>
<td>1410</td>
<td>1.14E-01</td>
</tr>
<tr>
<td>Flame 7</td>
<td>0.54</td>
<td>1696</td>
<td>8.47E-02</td>
</tr>
<tr>
<td>Flame 8</td>
<td>0.68</td>
<td>1945</td>
<td>5.66E-02</td>
</tr>
<tr>
<td>Flame 9</td>
<td>0.82</td>
<td>2156</td>
<td>3.03E-02</td>
</tr>
<tr>
<td>Flame 10</td>
<td>0.95</td>
<td>2314</td>
<td>8.94E-03</td>
</tr>
<tr>
<td>Flame 11</td>
<td>1.02</td>
<td>2355</td>
<td>2.93E-03</td>
</tr>
<tr>
<td>Flame 12</td>
<td>1.09</td>
<td>2361</td>
<td>8.20E-04</td>
</tr>
<tr>
<td>Flame 13</td>
<td>1.16</td>
<td>2346</td>
<td>2.70E-04</td>
</tr>
<tr>
<td>Flame 14</td>
<td>1.22</td>
<td>2322</td>
<td>1.10E-04</td>
</tr>
<tr>
<td>Flame 15</td>
<td>1.50</td>
<td>2212</td>
<td>1.00E-05</td>
</tr>
<tr>
<td>Flame 16</td>
<td>1.70</td>
<td>2133</td>
<td>0.00E+00</td>
</tr>
<tr>
<td>Flame 17</td>
<td>2.04</td>
<td>2012</td>
<td>0.00E+00</td>
</tr>
</tbody>
</table>

Table 5.1: CEA calculations for near-adiabatic calibration flames. Shown are the calculated temperatures (35 K below the adiabatic flame temperature) and the corresponding mole fractions of major species in the flat flames.
5.1 Procedure

For the initial calibration, room-temperature air (297 K) is used to set the nitrogen and oxygen calibration constants. The values of $C_{N_2}$ and $C_{O_2}$ are set to give a 3.76 molar ratio when performing the matrix inversion (in air) with all other species calibration constants set to zero. The Rayleigh temperature ($C_{T_{Rayleigh}}$) and Raman temperature ($C_{T_{Raman}}$) constants are then adjusted so that both temperatures equal 297 K. Figure 5.1 shows example mole fraction, temperature, and nitrogen-to-oxygen ratio profiles from a single 10-kHz burst acquired with the HEPBLS. As shown in Figure 5.1, each sub-figure displays an ensemble of 200 individual profiles, each corresponding to an instantaneous 1D image.

After the calibration constants in air have been determined, an unlit hydrogen/air flow corresponding to the composition of Flame 16 is used to adjust $C_{H_2}$. The results of this implementation within the non-reacting $H_2$/air flow are shown in Figure 5.2. It should be noted that the ratio of the Rayleigh temperature to the Raman temperature stays constant near unity and there is little, if any, shift in the N/O ratio. While there is some crosstalk from the hydrogen rotational lines onto the oxygen channel, it is much more significant at high temperatures, where there are higher populations of elevated rotational bands. For this reason, $C_{O_2} \ll H_2$ is more suitably determined via calibration under flame conditions.
Figure 5.1: Matrix inversion results in air at 297 K. Shown are the mole fractions of CO\textsubscript{2}, O\textsubscript{2}, N\textsubscript{2}, CH\textsubscript{4}, H\textsubscript{2}O, and H\textsubscript{2}, temperature, and the calculated background signal on the B3 and F560 channels. Temperature is reported as determined by the Rayleigh signal as well as the perfect gas temperature derived from the species concentrations (Raman). Ratios of the two temperatures as well as the nitrogen to oxygen are plotted. Data was acquired at 10 kHz using the HEPBLS.
Figure 5.2: Matrix inversion results in a non-reacting H₂/air flow at 297K. Shown are the mole fractions of CO₂, O₂, N₂, CH₄, H₂O, and H₂, temperature, and the calculated background signal on the B3 and F560 channels. Temperature is reported as determined by the Rayleigh signal as well as the perfect gas temperature derived from the species concentrations (Raman). Ratios of the two temperatures as well as the nitrogen to oxygen are plotted. Data was acquired at 10 kHz using the HEPBLS.

The remainder of the calibration constants must be determined under flame conditions either because their values change significantly at elevated temperature (\(C_{O₂H₂}, C_{H₂OH₂}\)) or they are present primarily in the combustion products.
In this work, the calibration constants are determined by the user as previously done [47], [73], [136] although a new automated, optimization method has been developed at Sandia National Laboratories which increases speed, accuracy, and daily precision [137]. For initial flame calibrations, iterations in the matrix inversion code were disabled and the Rayleigh temperature was set to be the “expected” temperature (35 K below adiabatic). Flame 11 was used to determine $C_{H_2O}$, as it is closest of the calibration flames to stoichiometric conditions, where peak temperatures and H$_2$O concentration occur. The constant describing the crosstalk of water onto the nitrogen channel $C_{N_2\ll H_2O}$ also was adjusted to bring the calculated Raman temperature much closer to the expected flame temperature value calculated from CEA. Flame 6 ($\phi = 0.41$) was used to determine $C_{O_2\ll H_2O}$ in order to account for excess oxygen due to the crosstalk from the water channel onto the oxygen channel and to determine $C_{H_2\ll H_2O}$ in order to account for crosstalk from the water channel onto the hydrogen channel. The value of $C_{H_2\ll H_2O}$ was set such that the hydrogen concentration was zero. Flame 16 ($\phi = 1.7$) was used to adjust for the effects of the rotational hydrogen lines on the oxygen channel ($C_{O_2\ll H_2}$) and the water channel ($C_{H_2O\ll H_2}$) respectively. The temperature and species concentration results from four of the calibration flames listed in Table 5.1 are shown in Figure 5.3 as a function of equivalence ratio ($\phi$). The solid red symbols represent the average value from 150 images acquired at 10 kHz using the HEPBLS. The solid lines represent the non-adiabatic, equilibrium calculations from CEA. For the first iteration of calibrations, the equivalence ratio shown in Figure 5.3 was determined from the CEA calculations, rather than derived from the species results.
Figure 5.3: Species concentrations and temperature in H$_2$/Air calibration flames as a function of equivalence ratio after a first iteration of calibrations. Note that the equivalence ratio plotted is an expected value calculated by the CEA code. Data was taken at 10 kHz with the HEPBLS.
After the initial iteration of calibrations, as shown in Figure 5.3, the calibration constants are fine-tuned in order to best fit the overall shape of the scalar dependence on equivalence ratio. In the example data presented, the species concentrations of water and nitrogen were very close to the expected values as determined from the CEA calculations, but both the oxygen and hydrogen values are negative. This is the result of too much crosstalk from the water channel; that is, $C_{O_2} ≪ H_2O$ and $C_{H_2} ≪ H_2O$ should be reduced. These calibration constants are further adjusted to give near-zero values of $O_2$ and $H_2$ at $\Phi=1$. The results from this update are shown in Figure 5.4.

Figure 5.4 shows that the value of oxygen is too high in fuel-rich flames as compared to the expected value of zero concentration. In order to drive the oxygen concentration values to zero under rich conditions, $C_{O_2} ≪ H_2$ was increased. However, Figure 5.4 shows that $O_2$ was calculated as near-zero in Flame 14 ($\Phi=1.225$), thus any increase $C_{O_2} ≪ H_2$ would result in negative $O_2$ concentrations in Flame 14. In this manner, $C_{O_2} ≪ H_2O$ also was adjusted. The value of the water calibration constant also was increased such that the derived concentrations matched the near-adiabatic calculations. Figure 5.5 displays the results after a third iteration on the calibration constants, where the equivalence ratio ($\phi$) was now calculated from species concentrations, rather than the calculated CEA values.
Figure 5.4: Species concentrations and temperature in H₂/Air calibration flames as a function of equivalence ratio after a second iteration of calibrations. Note that the equivalence ratio plotted is an expected value calculated by the CEA code. Data was taken at 10 kHz with the HEPBLS.
Figure 5.5: Species concentrations and temperature in H\textsubscript{2}/Air calibration flames as a function of equivalence ratio after a third iteration of calibrations. Note that the equivalence ratio plotted is based on species concentrations. Data was taken at 10 kHz with the HEBLs.
In Figure 5.5, it is noted that while the species concentrations agree with the flame calculations (expected values) from CEA, the temperature measurements are slightly too high. This may be due to slight nonlinearities in the camera over the dynamic range, but is most likely due to signal drift over time in addition to compounded errors arising from updating a large matrix of calibration constants to match five scalars over four flame conditions. In a final round of calibrations, the temperature constants were adjusted to give peak flame temperatures closer to the expected peak temperatures from the CEA predictions. Changing the temperatures affects all species, as their values are dependent upon the Sandia polynomials and the Ramses calculations, both of which are highly temperature dependent. The results of the final adjustments for the 10-kHz example test case are shown in Figure 5.6, where the set of species-specific signal and crosstalk calibration constants are set to achieve agreement between the calibration-flame measurements and the expected values over the entire range of conditions.

In principle, the Raman-response matrix coefficients can be updated such that any specific flame condition will match the expected values as determined from the CEA calculations; however, this does not ensure that other flame conditions will match. The reported procedure and final set of calibration constants are determined to minimize error over all calibration conditions. Figure 5.6 demonstrates this principle as there is very good agreement between the four species and Rayleigh-derived temperature across a broad range of conditions. After the final round of iteration, which target flame conditions, 10-kHz data was acquired in air and processed again. After the iterative procedure to determine the calibration constants (in flames), the predicted temperatures
were $T_{\text{ray}}=294$ K and $T_{\text{ran}}=298$ K as compared to the expected value of $T = 297$ K. These derived temperature values are well within the accuracy of the measurements discussed below and demonstrate minimal introduction of error at lower temperature when calibrating at flame conditions. While it was fortuitous that the room-temperature measurements agreed with expected values, it should be noted that if it is necessary to choose between accuracy at high temperatures and accuracy in cold air, flame temperatures are favored, as these conditions are the main area of interest of this research. Finally, it is noted that once the calibration constants are determined in the laminar flames (Table 5.1), they are not adjusted further for the turbulent flame measurements.
Figure 5.6: Species concentrations and temperature in H$_2$/Air calibration flames as a function of equivalence ratio after a fourth iteration of calibrations. Note that the equivalence ratio plotted is based on measured species concentrations. Data was taken at 10 kHz with the HEPBLS.
5.2 Data Processing Verification

As a verification of the calibration procedure discussed in Section 5.1 and the matrix inversion data reduction code and procedure developed at Ohio State, a set of near-adiabatic (“Hencken”) flame data was taken using the Raman/Rayleigh scattering setup at the Combustion Research Facility (CRF) at Sandia National Laboratories. The experimental details of the CRF experimental setup has been described in detail in Refs. [37], [48], [68], [141]. The Rayleigh/Raman scattering system at the CRF can be considered as the leading facility for acquiring such measurements and the data used to assess the matrix inversion processing was taken with a state-of-the-art imaging system consisting of cryogenically-cooled, ultra-low noise CCD cameras and multiple Nd:YAG lasers yielding single pulse energies of more than 900 mJ. The data acquired at the CRF is not meant as a direct comparison to the high-speed data reported in this dissertation, but is used as a means to test the data processing of flame data with high signal-to-noise ratios from a proven Raman/Rayleigh test facility. The calibration-flame measurements (from the 14 flames listed in Table 5.1) taken at the CRF at Sandia National Laboratories were performed with the same Hencken burner used for the calibration of the high-speed measurements performed at OSU. The results from 100 single-shot measurements at each flame condition are shown in Figure 5.7. Also shown in Figure 5.7 are the near-adiabatic flame calculations from the CEA code, plotted as a solid black line. As shown in Figure 5.7, the derived scalar values exhibit little scatter around the mean value (for each flame condition), which agrees well with the expected value from the CEA calculation. Signal-to-noise ratios for the CRF data are shown in Table 5.2. The SNR of the scalar value,
which is a function of the measurement and the data processing, is consistent with previously-reported values [136], [142]. Thus, it is concluded that the overall calibration and matrix inversion processing methodology applied at OSU (and subsequently to the acquired high-speed data) works as expected.

\[
\begin{array}{lcccc}
 & H_2/air Flame & & H_2/air Flame & \\
 & \phi=0.27 & & \phi=2.04 & \\
X_{H_2} & 206 & & & \\
X_{O_2} & 49 & & & \\
X_{H_2} & & 37 & & \\
X_{H_2O} & & & 57 & \\
T_{ray} & 123 & & & \\
\text{phi} & 43 & & & \\
\end{array}
\]

Table 5.2: Representative signal-to-noise ratios in near-adiabatic calibration flames from measurements taken at the CRF at Sandia National Laboratories. Signal-to-noise ratios taken in H\textsubscript{2}/air Hencken flames with 60 \(\mu\)m resolution are averaged over the entire probe volume (3.6 mm) and 91 shots.
Figure 5.7: Measured temperature and mole fractions in near-adiabatic flat flames at the Combustion Research Facility (CRF) at Sandia National Laboratories. Data taken at the CRF used the same OSU Hencken burner used for calibration at Ohio State. Measured data is plotted as blue symbols and the CEA calculations are shown as the solid black line.
5.3 Calibration Results at Low Repetition Rates (10 Hz)

As noted in Section 3.2, measurements were acquired at low repetition rates (10 Hz) using the high-speed imaging system in conjunction with a commercially-available Q-switched, Nd:YAG laser (Spectra Physics Pro-290). The goal of these measurements was to isolate the effect of the imaging system (as limited by a high-speed, intensified CMOS camera for the Raman measurements) without the influence of the custom HEPBLS.

Specifically, the 10-Hz, Q-switched, Nd:YAG exhibits low pulse-to-pulse energy fluctuations (< 2 % RMS), high beam quality ($M_2 < 1.2$), low beam divergence (<0.5 mRad), and high beam-pointing stability. In terms of photon generation and without regards to repetition rate, the 10-Hz, Q-switched, Nd:YAG represents an excellent laser source for Raman/Rayleigh measurements and is used for two purposes: (i) to assess the limitations of the data due to the high-speed detection equipment and (ii) generate a baseline data case for comparison to high-speed data acquired using the custom HEPBLS laser. Shown in Figure 5.8 are the species mole fraction and temperature results using the 10-Hz laser system for the near-adiabatic hydrogen/air flames described in Table 5.1. For all results shown in Figure 5.8, the CEA calculations are plotted as solid black lines. The left-hand column shows the mean species mole fractions and temperatures averaged over the entire 6.1-mm line segment from 151 images with average laser energies of 350 mJ per pulse. The plotted error bars correspond to $2\sigma$ in the species mole fraction (vertical) and calculated equivalence ratio determined from the species mole fractions (horizontal), where $\sigma$ is the standard deviation of the measurement. It is noted that these error bars describe the measurement precision and do not include any uncertainty due to calibration.
procedure discussed in Section 5.1. Figure 5.8 shows that the mean value of the species mole fractions and temperature align very well with the expected values from the near-adiabatic, equilibrium CEA calculations.

Scatter plots of the scalars are shown in the right-hand column of Figure 5.8. The scatter plots, which are created by plotting every 160-µm pixel for all 150 images (~80,000 data points), show that the majority of the data points follow closely to the near-adiabatic flame curves, particularly at low equivalence ratios. However, it is observed that there are notable instances of non-physical negative values from hydrogen and oxygen near $\phi = 1$ and positive hydrogen mole fractions under lean flame conditions and positive oxygen mole fractions under rich flame conditions, where each are expected to be zero. These values lead to slightly positive mean values for $O_2$ and $H_2$ under rich and lean conditions, respectively. Much of the scatter under rich conditions is caused by uncertainty in the oxygen mole fraction due to crosstalk from hydrogen and water. Since the equivalence ratio is calculated from species concentrations and is very sensitive to the oxygen level, negative oxygen values lead to artificially high equivalence ratios and large uncertainty in the derived equivalence ratios. This is noted by the large horizontal error bars on the mean values (left column, Figure 5.8) at high equivalence ratios.

As a measure of the expected precision of the system, signal-to-noise ratios (SNR) for the 10-Hz data are reported in Table 5.3 for certain operating conditions. Here the SNR is defined as the mean signal divided by the standard deviation for a particular flame operating condition; a convention established at the CRF at Sandia National Laboratories.
Figure 5.8: Measured temperature and mole fractions in near-adiabatic flat flames at 10 Hz. Shown in the left column are mean values (red symbols) for the temperature and mole fractions. The vertical error bars represent 2σ in the scalar values and the horizontal error bars represent 2σ in the calculated equivalence ratio. Scatter plots of the single-shot measurements are shown in the right column as solid red symbols. Also shown are the CEA calculations displayed as the solid black line.
Table 5.3: Representative signal-to-noise ratios in near-adiabatic calibration flames at 10 Hz. SNR is determined within the H₂/air Hencken flames from the entire probe volume (6.1 mm) and 150 individual laser shots. For the “cold” conditions, the SNR of N₂, O₂, and Tray were determined in air and for H₂ and phi, the SNR was determined in an unlit flow of H₂/air at φ=1.7 and 300K.

<table>
<thead>
<tr>
<th></th>
<th>Cold (300 K)</th>
<th>H₂/air Flame φ=0.27</th>
<th>H₂/air Flame φ=2.04</th>
</tr>
</thead>
<tbody>
<tr>
<td>X₅₂</td>
<td>120</td>
<td>60</td>
<td></td>
</tr>
<tr>
<td>X₂₀₂</td>
<td>33</td>
<td>16</td>
<td></td>
</tr>
<tr>
<td>X₂H₂</td>
<td>54</td>
<td>13</td>
<td></td>
</tr>
<tr>
<td>X₂H₂O</td>
<td></td>
<td></td>
<td>15</td>
</tr>
<tr>
<td>Tᵣay</td>
<td>261</td>
<td>114</td>
<td></td>
</tr>
<tr>
<td>phi</td>
<td>20</td>
<td>10</td>
<td></td>
</tr>
</tbody>
</table>

5.4 Calibration Results at 10 kHz

Figure 5.9 shows the species mole fraction and temperature results for the near-adiabatic hydrogen/air flames taken with the HEPBLS at a 10-kHz acquisition rate. For all results shown in Figure 5.9, the CEA calculations are plotted as solid black lines. The left-hand column shows the mean species mole fractions and temperatures averaged over the entire 6.1 mm line segment from 150 images with average laser energies of ~440 mJ per pulse. The plotted error bars correspond to 2σ in the species mole fraction (vertical) and calculated equivalence ratio (horizontal), where σ is the standard deviation of the measurement. Similar to the 10-Hz data reported above, these error bars only represent measurement precision and do not include systematic errors due to the calibration procedure. The right-hand column shows the scatter plots of the single-shot scalar measurements. The scatter plots are created by plotting the scalar values from every 160-μm pixel for 170 bursts of 150-images (~80,000 data points), where each image is spaced
by 100 μs. Similar to the 10-Hz data, Figure 5.9 shows that the majority of the data points follow closely to the near-adiabatic flame curves and mean species mole fractions and temperature align very well with the expected values from the CEA calculations. In addition, the scatter is visibly indistinguishable from the measurements performed at 10 Hz using the Q-switched, Nd:YAG laser. Also similar to the 10-Hz measurements are the observations of instances of non-physical negative values from hydrogen and oxygen near φ = 1 and positive hydrogen and oxygen mole fractions under lean and rich flame conditions, respectively, where each are expected to be zero.

Table 5.4 shows the signal-to-noise ratio (SNR) for the 10-kHz data under selected operating conditions. The SNR in Rayleigh temperature is reduced under low-temperature conditions (high signal) when using the HEPBLS as compared to the measurements at 10 Hz. This reduction in SNR under high-signal conditions is due to stronger pulse-to-pulse fluctuations in laser energy from the HEPBLS. However, as shown in Figure 5.10, the SNR of the temperature measurements are quite comparable between the 10-Hz and 10-kHz data at higher temperatures. Despite the reduction in SNR in the temperature measurement when using the HEPBLS, little effect is observed on the signal-to-noise ratios of the species data derived from the Raman measurements (X_{O_2}, X_{N_2}, X_{H_2O}, and X_{H_2}). Section 5.5 discusses this facet in more detail.
Figure 5.9: Measured temperature and mole fractions in near-adiabatic flat flames at 10 kHz. Shown in the left column are mean values (red symbols) for the temperature and mole fractions averaged over 150 images and 170 bursts along a 6.1 mm line segment, 18 mm above the surface of the burner. The vertical error bars represent 2$\sigma$ in the scalar values and the horizontal error bars represent 2$\sigma$ in the calculated equivalence ratio. Scatter plots of the single-shot measurements are shown in the right column. Also shown are the CEA calculations displayed as the solid black line.
<table>
<thead>
<tr>
<th></th>
<th>Cold” (300 K)</th>
<th>H₂/air Flame φ=0.27</th>
<th>H₂/air Flame φ=2.04</th>
</tr>
</thead>
<tbody>
<tr>
<td>$X_{N_2}$</td>
<td>123</td>
<td>67</td>
<td></td>
</tr>
<tr>
<td>$X_{O_2}$</td>
<td>33</td>
<td>17</td>
<td></td>
</tr>
<tr>
<td>$X_{H_2}$</td>
<td>54</td>
<td></td>
<td>15</td>
</tr>
<tr>
<td>$X_{H_2O}$</td>
<td></td>
<td></td>
<td>15</td>
</tr>
<tr>
<td>$T_{ray}$</td>
<td>110</td>
<td>50</td>
<td></td>
</tr>
<tr>
<td>phi</td>
<td>27</td>
<td>12</td>
<td></td>
</tr>
</tbody>
</table>

Table 5.4: Representative signal-to-noise ratios in near-adiabatic calibration flames at 10 kHz. SNR is determined within the H₂/air Hencken flames from the entire probe volume (6.1 mm) and 150 individual laser shots. For the “cold” conditions, the SNR of N₂, O₂, and Tray were determined in air and for H₂ and phi, the SNR was determined in an unlit flow of H₂/air at φ=1.7 and 300K.

5.5 Uncertainty Analysis and Discussion

Due to the fluctuating nature of turbulent flows, it is often difficult to directly determine uncertainty under turbulent flame conditions. If the measured scalars were simply functions of a series of independent variables written as:

$$\phi = \prod_{i=1}^{n} y_i^{c_i}$$  Equation 5.1

then the uncertainty ($\varepsilon_\phi$) in any scalar could be determined as:

$$\varepsilon_\phi = \left( \sum_{i=1}^{n} c_i (\varepsilon_{y_i})^2 \right)^{1/2}$$  Equation 5.2

where $\varepsilon_{y_i}$ represents the associated uncertainty in each quantity $y_i$. However the uncertainty estimation using Equation 5.2 only applies to the situation where the quantities $y_i$ are uncorrelated and their associated uncertainties are independent random variables [143]. As shown in Chapters 3 and 4, the local scalar value (species or
temperature) is correlated with all other scalars through the “crosstalk” parameters and the mixture-averaged Rayleigh scattering cross section. In this manner, a straightforward and explicit uncertainty analysis using Equation 5.2 is not possible.

Instead, it is convention to rely on measurements in stable, laminar, near-adiabatic flames [77], [131], [144], [145] to determine measurement accuracy and precision. In this work, the calibration measurements in air and the set of near-adiabatic flames described listed in Table 5.1 are used to estimate the uncertainty in the single-shot measurements within the turbulent flames. There are two components to the uncertainty of the turbulent-flame measurements described in the remaining portions of this dissertation: the precision (or the reproducibility or reliability of the measurement) and the accuracy (the closeness of the measurement to the true value). Measurement precision is estimated using the variance (or standard deviation) of the scalar measurements in stable laminar flames, as the precision of the measurements are closely related to the random error of the experiment [140]. This is not possible in turbulent flames because measurement fluctuations (or variance) include both the process variance (i.e., turbulent fluctuations) in addition to sources of random error that lead to imprecision. While the standard deviations of the laminar flame measurements are shown as “error bars” in the data reported in Figures 5.8 and 5.9, the signal-to-noise ratios (Figure 5.10), and probability density functions (Figure 5.14) for the 10-Hz and 10-kHz data are reported explicitly as function of equivalence ratio to highlight the measurement precision under presumed constant measurement conditions, i.e., laminar flames.
The SNR results from the 10-Hz data (shown in the left column of Figure 5.10) can be used to first examine the noise characteristics of the Rayleigh and Raman detectors used within the imaging spectrometer and the associated effects on the measurement imprecision. The 10-Hz measurements are used for this purpose because of the low pulse-to-pulse energy fluctuations (~1% RMS) and thus the relative insensitivity of the measurement SNR to accurate laser-energy-fluctuation corrections. Figure 5.10 shows that the measured SNR in the laminar flames decreases with increasing equivalence ratio for the Rayleigh temperature, $N_2$, and $O_2$ (decreasing number density), while the SNR increases with increasing equivalence ratio for $H_2$ and $H_2O$ (increasing number density), both of which are the expected trends.

While the results of Figure 5.10 show the measurement SNR for a given flame condition (i.e., local mixture fraction in the turbulent flames), it does not give insight into the dominant sources of noise or measurement limitations for the scalar results. To examine this, the SNR of the measured signal (following laser energy fluctuation corrections, but before matrix inversion processing) should be examined. Following the nomenclature of Clemens [42], the SNR of the measurement (in terms of photons per pixel or signal, $S_{pp}$) without consideration of imprecision due to laser energy corrections or random error arising during processing is written as:

$$\text{SNR} = \frac{\eta S_{pp}G}{(K\eta S_{pp}G^2 + N_{\text{cam}}^2)^{1/2}}$$  \hspace{1cm} \text{Equation 5.3}$$

where $(K\eta S_{pp}G^2)^{1/2}$ is the “shot noise” arising from the statistical fluctuations of the number of photoelectrons generated at each pixel, $K$ is a noise factor that quantifies noise generated through the overall gain process between a photocathode (if present) and
sensor array, $G$ is the overall electron gain from the photocathode (if present) to the sensor, $\eta$ is the quantum efficiency, and $N_{\text{cam}}$ represents the intrinsic noise of the camera, which includes additional amplifier noise, thermal and dark-current noise, and most notably, the “read noise”, which describes the quantization noise in the A/D conversion of the signal. For an un-intensified camera, $K = G = 1$. Equation 5.3 reveals many important facets about the effects of the detector on the measurement SNR. First it is noted that if $(KS_eG)^{1/2} \gg N_{\text{cam}}$, the dominant noise source is the photoelectron shot noise and the SNR will increase as approximately $S_e^{1/2}$. If $N_{\text{cam}} \gg (KS_eG)^{1/2}$, then the SNR will increase as approximately $S_e$. Another implication from Equation 5.3 is the fact that although intensified cameras exhibit high levels of “noise” as compared to their un-intensified counterparts, they can still be operated in the shot-noise limited regime by increasing the gain until $(KS_eG)^{1/2} \gg N_{\text{cam}}$. However, increasing the gain ($G$) decreases the number of photoelectrons that can be captured per pixel ($S_{pp}$) before saturation since $S_e \sim S_{pp}G$. In this manner, increasing the gain allows shot-noise limited behavior, but limits SNR due to a limited number of collected photoelectrons.
Figure 5.10: Signal-to-noise ratios of species mole fractions in near-adiabatic, laminar flames as a function of equivalence ratio. Results for the 10-Hz data are shown in the left-hand column and results for the 10-kHz data taken with the HEPBLS are shown on the right.
Figure 5.11 shows the SNR of the measured signal (following laser energy fluctuation corrections, but before matrix inversion processing) at each one of the flame conditions reported in Figure 5.10. The SNR of the measured signals are shown as blue symbols and the SNR of the deduced temperature or species mole fraction (reproduced from Figure 5.10) at the corresponding signal level is shown as a solid red symbol. By comparing the two SNR values, the effects of the detector and the processing can be separated to some degree. Here it is seen that the SNR of the processed mole fractions are sometimes higher than that of measured signal, particularly in the 10 kHz data where there are strong shot-to-shot pulse energy fluctuations. This phenomenon stems from the fact that the processed species mole fraction measurements are highly coupled. The species mole fractions are deduced from the individual species concentration divided by the total concentration, creating a dampening effect on noise, i.e., the effect of noise largely “cancels out”. When the measured signal is lower than expected due to a large drop in laser energy, it affects all the Raman signals equally and, therefore, the total concentration as well. This drop in signal is somewhat negated when the individual species concentration is divided by the total concentration, leading to a higher SNR in the processed data. Figure 5.12 shows the same SNR of the measured signal (following laser energy fluctuation corrections, but before matrix inversion processing) along with the SNR from the calculated species concentrations (red). For the 10 Hz data, the SNR of the measured signal is always greater than that of the calculated species concentration and temperature. This is not the case, however for the results at 10 kHz. While the SNR of the measured Rayleigh signal is higher than that of the calculated temperature, the SNR of
the calculated species concentrations are larger than their respective measured signals at 10 kHz. Because of the highly coupled nature of the measurements, it is likely that the relatively high SNR of the Rayleigh temperature measurement is increasing the SNR of the species concentration measurements. This suggests that the effects of the pulse-to-pulse fluctuations from the HEPBLS have been better corrected in the CMOS-only Rayleigh measurements than in the high-speed IRO Raman measurements. It also is a possibility that there is some non-linearity in the IRO response when such large shot-to-shot signal fluctuations occur.
Figure 5.11: Signal-to-noise ratios of species mole fractions in near-adiabatic, laminar flames as a function of equivalence ratio. Results for the 10-Hz data are shown in the left-hand column and results for the 10-kHz data taken with the HEPBLS are shown on the right. Results for processed data are plotted in red and results for measured signal (following laser energy fluctuation corrections, but before matrix inversion processing) are plotted in blue.
Figure 5.12: Signal-to-noise ratios of species concentrations in near-adiabatic, laminar flames as a function of equivalence ratio. Results for the 10-Hz data are shown in the left-hand column and results for the 10-kHz data taken with the HEPBLS are shown on the right. Results for processed data are plotted in red and results for measured signal (following laser energy fluctuation corrections, but before matrix inversion processing) are plotted in blue.
In order to better illustrate the noise-characteristics of the detection system for each camera, the detector noise ($N_{\text{cam}}$) was calculated for each measurement case (10-Hz measured signal, 10-Hz processed data, 10-kHz measured signal, 10-kHz processed data) based on the most fuel-rich flame condition. The expected SNR shot-noise dependence from Equation 5.3 is plotted in Figure 5.13. For the Rayleigh (CMOS-only) conditions (Figure 5.13, top), the 10-Hz data closely follows the expected SNR dependence for measurements in the shot-noise regime. For measurements at 10 kHz, both the raw signal and processed data follow shot-noise limited characteristics under rich conditions (low signal levels), but exhibits departure from the not-noise-limited regime for high-signal conditions. This suggests the SNR goes from being shot-noise dependent to being limited by the laser-energy correction as signals increase. The process was repeated for the HS-IRO/CMOS data using the N$_2$ mole fraction as an example (Figure 5.13 bottom) and the same trend is observed. The processed N$_2$ mole fraction data (both at 10 Hz and 10 kHz) appears to be shot-noise limited, suggesting that the data processing has a limited ability to filter out the energy-correction error in low SNR measurements.

An additional way to view the scatter of the laminar flame measurements is through the use of probability density functions (pdfs) of scalar measurements. The 10-Hz and 10-kHz measurements both are plotted in Figure 5.14. It is important to note that small shifts in the pdf in terms of the scalar values are due predominately to the chosen pdf bin sizes and corresponding boundaries. For example, assume two temperature measurements of 1974 K and 1975 K for the 10-Hz and 10-kHz measurements, respectively. The difference between these two temperature results is not statistically
significant; however, they would be binned separately, possibly creating a small shift between the reported pdfs of the two repetition rates. However, the peak value and overall shape should be less sensitive to the chosen pdf bin sizes and associated boundary effects.

While the comparison between the 10-Hz and 10-kHz measurements has been discussed previously in qualitative terms, the pdfs shown in Figure 5.14 compare the two data sets quantitatively. More specifically, the comparison of pdfs allow the direct observations in differences in accuracy (closeness to mean value) and precision (SNR value) between the two data sets, where the only difference between the measurement campaign involved the laser source. Figure 5.14 shows that with increasing temperature, the pdfs of the temperature become very similar, re-enforcing the fact that for high number densities (lower temperatures), the limiting factor in the SNR is the correction for pulse-to-pulse energy fluctuations when using the HEPBLS, but at lower number densities (or higher temperatures and lower signals and SNR), the limiting factor in terms of the SNR is detector noise. For all species, the pdfs are comparable between the two measurement campaigns over all flame conditions, with small differences associated with binning boundaries as discussed above. This implies that the increased uncertainty in the temperature measurements at 10 kHz (as compared to 10 Hz) has little effect on the derived species mole fractions.
Figure 5.13: Signal-to-noise ratio as a function of equivalence ratio for Rayleigh temperature (top) and nitrogen mole fraction (bottom). Also plotted are the shot-noise limits calculated with Equation 5.3.
The accuracy, defined as the difference between the measured mean values and the expected or “true” mean values, of the turbulent measurements also can be estimated using laminar flame data, though this requires assuming that the actual temperatures and species concentrations can be accurately described by the near-adiabatic flame.
calculations. In this work, it is assumed that the “true” temperatures are 35 K below adiabatic to account for heat loss to the burner and radiation effects [63], [140]. In general, the measurement accuracy is limited by calibration quality (including uncertainty in crosstalk and background corrections), laser-alignment drift, beam steering, flow-rate stability (particularly downstream where room-air currents can influence flame position), ambient temperature fluctuations (a substantial issue when operating the HEPBLS), and fluorescence from interfering species. Of these factors, beam steering due to fluctuating refractive index gradients and fluorescence are the only two unique to turbulent flame conditions. Beam steering is small compared to the size of the software bins in these flames and the majority of 532-nm Raman interference comes from C₂, which is not present within the hydrogen flames considered in this work. Measurement accuracy was estimated by comparing the mean value of the measured scalars to the near-adiabatic calculations. For conditions in which the measurements and the calculations do not occur at the same equivalence ratio, interpolation of the scalar values was performed based on the calculations at the nearest equivalence ratios. The absolute value of the difference between measurements and the near-adiabatic calculations was normalized by the measured mean scalar value to determine a percentage inaccuracy. These estimated inaccuracies are shown in Figure 5.15. The inaccuracies are plotted on a semi-log scale due to the fact that as the mean value approaches zero, the estimated inaccuracy approaches infinity. In general, most scalars are accurate to within 2-6% where sufficient concentrations exist for meaningful measurement. The reported inaccuracies can then be
combined with the measured imprecision to determine an overall uncertainty in the turbulent flame measurements, which depend on the local flame conditions.
Figure 5.15: Estimated inaccuracies in near-adiabatic flames
Chapter 6: Characterization of Turbulent Flame Measurements

In this chapter, measurements and derived statistics from turbulent H\textsubscript{2}/N\textsubscript{2} flames using the current high-speed Raman/Rayleigh imaging system are compared to previously reported results from DLR available within the TNF database [9], [24] to assess the accuracy of single-shot Raman/Rayleigh scattering measurements under turbulent flame conditions. First, the scalar means, scalar RMS fluctuations, and scalar means conditioned on mixture fraction derived from the 10-Hz measurements are compared to the previous measurements from DLR as a method to validate our “single-shot” measurement capability without the added complexity of the HEPBLS. Subsequently, the comparison is repeated with the 10-kHz measurements (approximately 180 20-ms bursts) acquired utilizing the HEPBLS.

The previous single-point measurements at DLR utilized a flashlamp-pumped dye laser operating at 488nm to excite the Raman bands at a repetition rate of 5 Hz. The laser pulse was retro-reflected with a spherical mirror back through the turbulent flame to give effective pulse energies of up to 4.5 J/pulse with effective beam diameters of approximately 400 \( \mu \text{m} \) (FWHM) or 650 \( \mu \text{m} \) (1/e\(^2\)). A spectrograph was used to separate and disperse the Raman signals (O\textsubscript{2}, N\textsubscript{2}, background, H\textsubscript{2}O, and H\textsubscript{2}) onto individual photomultiplier tubes (Hamamatsu R928 or R3896). Data reduction was performed with the matrix inversion method using calibration data collected in steady, laminar flat flames.
stabilized above the surface of a McKenna burner. For the data presented here, 300 single-pulse measurements are used, which have reported standard deviations from the flat-flame measurements on the order of 1-5% for the individual scalars. The uncertainty in the scalars is estimated to be ~1% for N\textsubscript{2} and H\textsubscript{2}O and 3-5% for O\textsubscript{2} and H\textsubscript{2} [9].

6.1 Comparison of 10-Hz Measurements with Previous Results

Figure 6.1 shows the mean and RMS fluctuations of species mole fractions and temperature (K) as a function of radial distance (mm) for the measurements acquired at 10 Hz in the H3 flames as described in Chapter 3. The solid red symbols show the mean value of the species mole fractions and temperature from 150 images averaged over a 160-\(\mu\)m radial distance (38 in total). The measurements were acquired at 10 Hz using a commercial Q-switched Nd:YAG laser (Spectra Physics Pro 290) at 532 nm with average laser pulse energies of 350 mJ. The error bars, plotted only on the center radial position, correspond to 2\(\sigma\) from the laminar calibration flame results averaged over all equivalence ratios. The open red symbols correspond to the RMS fluctuations of the scalar measurements. The data plotted as blue symbols correspond to the species mole fractions and temperature measurements from DLR [2] available through [9], [24]. The solid blue symbols correspond to the mean from 300 measurements at radial spacing of approximately 1.5 mm. Each data point corresponds to the mean scalar value averaged over a 400-\(\mu\)m volume as defined by the spatial resolution. The DLR RMS values are shown as open blue symbols.
Figure 6.1 shows that, overall, the mean and RMS values of the current measurements agree well with the previous data taken at the DLR. As expected from the calibration results shown in Chapter 5, there is some difficulty measuring small oxygen mole fractions approaching zero. This is apparent in the data reported at x/D=10, which is highlighted in Figure 6.2 and shows that there are small negative values of oxygen mole fraction measured near the centerline. While these values are within the expected error of the measurement (here only precision error is plotted), they are likely to cause errors in the mixture fraction measurements.
Figure 6.1: Radial profiles of the mean and RMS fluctuations of major species mole fraction and temperature in the H3 flame. Current measurements from the OSU high-speed Raman system at 10 Hz are plotted in red and measurements taken at DLR are plotted in blue. The solid markers represent the mean, while the open symbols represent the RMS fluctuations.
Figure 6.2: Radial profile of the mean and RMS fluctuations of $O_2$ mole fraction at $x/D=10$. Current measurements from the OSU high-speed Raman system at 10 Hz are plotted in red and measurements taken at DLR are plotted in blue. The solid markers represent the mean, while open symbols represent the RMS fluctuations.

Figure 6.3 shows “scatterplots” of the measured scalars (red symbols) as a function of mixture fraction at $x/D = 10$, 20, and 40 and sampled from radial positions separated by 2-mm increments. The black line shows the adiabatic, equilibrium solution of the scalars as a function of mixture fraction. Overall, the instantaneous data follows the adiabatic, equilibrium curves as expected with few samples indicating extinction. However, small errors in measuring small values of the species are noted in Figure 6.3, especially at $x/D = 40$. For example, at $x/D = 40$, it is seen that many of the individual oxygen measurements are small negative values. At this axial position, the hydrogen measurements also are negative near the stoichiometric mixture fraction. These negative values influence the deduced mixture fraction measurement by skewing it either too lean...
or two rich. At $x/D = 40$ the small negative values of hydrogen near the stoichiometric mixture fraction skew the mixture fraction (calculated from the species concentrations) towards the lean side of the flame ($\xi < \xi_s$), causing a shift in the scatter in all the measured scalars at the stoichiometric mixture fraction. These slightly negative values are the result of trying to achieve the best overall fit for a broad range of conditions in the laminar calibration flames and are reflective of the noise characteristics of the intensified CMOS camera system.

Figure 6.4 shows the conditionally-averaged values of the scalars as a function of mixture fraction. Measurements are from axial positions of $x/D = 10$, 20, and 40 and from radial positions separated by 2-mm increments. Measurements at a given axial position were combined and separated into 0.05-wide mixture fraction bins. Each bin with at least 0.5% of the total number of measurement samples was averaged and plotted as a function of the mean mixture fraction of a given bin. As before, the red symbols correspond to the current data taken at 10 Hz at Ohio State while the blue symbols correspond to data taken at the DLR. Also shown in Figure 6.4 are the adiabatic, equilibrium flame solutions for each scalar, plotted as a solid black line.

At axial positions of $x/D=10$ and $x/D=20$ all mixture fraction bins (0 to 1) are filled, which is consistent with measurements between the nozzle and flame tip where all levels of mixed fluid are present along with pockets of unmixed and unburned gases. For measurements downstream at $x/D=40$, which is beyond the flame tip, only the bins between a mixture fraction of zero (pure air) and the stoichiometric mixture fraction remain populated.
For axial positions $x/D=10$ and $x/D=20$, the current data matches both the adiabatic flame curves and the DLR data very well. For the $x/D=10$ temperature and water measurements, there is some deviation from the adiabatic flame curve, although this feature is consistent between the current data and the DLR data. As seen in both Figures 6.2 and 6.3, the current data contains a finite number of samples with small negative values of oxygen. These negative values skew the calculated mixture fraction towards rich conditions away from the adiabatic flame curve. It should be noted that the DLR data filters out negative values, which can lead to an observed shift between DLR data and data taken with other systems [8].

At $x/D=40$ (Figure 6.1), this deviation becomes clear, where the deviation from the adiabatic flame curve (and the DLR data) is seen in all scalars near the stoichiometric mixture fraction. While these measurements are statistically significant (~2% of the flame data), their deviation from previously reported data is likely due to the negative values of both hydrogen and oxygen measured at $x/D=40$ (see Figure 6.3). These negative values exist where water is at its peak, suggesting an overcorrection for water crosstalk at high temperatures in the calibration. Since $x/D=40$ is beyond the flame tip, few mixture fraction values above stoichiometric are seen in either data set.
Figure 6.3: Scatterplots of the major species mole fraction and temperature in mixture fraction space for the H3 flame. Measurements are shown for x/D = 10, 20, 40. Current measurements from the high-speed Raman system at 10 Hz are plotted as red symbols and the adiabatic flame conditions plotted as black lines.
Figure 6.4: Conditionally-averaged values of major species mole fraction and temperature in the H3 flame. Measurements are shown for \( x/D = 10, 20, 40 \). Current measurements from the high-speed Raman system at 10 Hz are plotted in red and measurements taken at DLR plotted in blue. The single-shot results are conditionally averaged within distinct mixture fraction intervals of 0.5 and plotting is limited to bins with samples containing more than 0.5% of the total number of measurement samples.

The adiabatic flame conditions are shown as black lines.
In order to take a closer look at the comparison between the current data and the previous DLR data, probability density functions (PDFs) of the scalar values for selected mixture fraction bins were calculated and shown in Figure 6.4. The results for a stoichiometric bin, (ξ=0.275-.3025), a lean bin (ξ=0.075-0.125), and a rich bin (ξ=0.475-0.525) at x/D=20 are shown in Figures 6.5, 6.6, and 6.7, respectively. Figure 6.5 shows that, overall, the PDFs of the current 10-Hz measurements and the previous DLR measurements are very similar at x/D = 20, with small differences attributed to increased noise levels in the present experiment. The increased noise can lead to an increased standard deviation for a given scalar, and since mixture fraction is a derived quantity from the major species, noise also can lead to errors in mixture fraction, which can in turn, lead to a biasing of the PDFs for a given mixture fraction bin. Specifically, with the exception of nitrogen and water mole fractions, the two measurement sets have very similar means and standard deviations for the stoichiometric bin. Both nitrogen and water have a consistent mean between the two data sets, but the current 10-Hz measurements show a slight broadening (increase in standard deviation) of the PDF.

For the selected lean mixture fraction bin (Figure 6.6), there is good agreement between the current 10-Hz results and the previous DLR data for all scalars. Considering that the hydrogen mole fraction has a high probability of a value of zero (which is a challenge for the current high-speed imaging system), the agreement between the two data sets is encouraging. For the rich conditions shown in Figure 6.7, the PDFs show consistent mean values but a higher standard deviation for the current measurements as
compared to the previous DLR data for all scalars. This is most likely due to the
difficulty in measuring zero values of the oxygen mole fraction.

The PDFs of the stoichiometric bin at $x/D=40$ are shown in Figure 6.8. For these
conditions, the PDF of the temperature has a similar standard deviation, but there is a
shift in the mean value towards higher temperatures. The species measurements show
both increased standard deviation and a shift in the mean values for the current data as
compared to the previous DLR data. It is important to note that at $x/D=40$, the
measurements are at a location that is beyond the average position of the flame tip, so
water should be present at all measurement locations. For the selected stoichiometric
mixture fraction bin, the mean value of both the oxygen and hydrogen mole fraction is
slightly negative. An overcorrection of water crosstalk leads to negative values for both
hydrogen and oxygen mole fractions and subsequent errors in the deduced mixture
fraction. The sensitivity of the species to the water crosstalk not only leads to effective
shifts in the mean values but to a higher standard deviation as well. In addition, the higher
standard deviation of the PDFs of the current 10-Hz mole fraction measurements also are
due to lower SNR values at higher mixture fractions (similar to what was observed in the
SNR data at high equivalence ratios in Chapter 5). The fact that the departures of the
current PDFs from the PDFs of the DLR data is more pronounced at $x/D=40$ as compared
to $x/D = 20$ could be due shifts in the calibrations over time or interference from small
levels of dust and particulate entrained at $x/D = 40$. 

146
Figure 6.5: Probability density functions of temperature and species mole fractions near stoichiometric conditions at x/D=20. Plotted are the PDFs of the current 10-Hz data (red) and previous DLR data (blue) for the stoichiometric mixture fraction bin (ζ=0.275-0.325).
Figure 6.6: Probability density functions of temperature and species mole fractions for lean mixture fraction conditions at x/D=20. Plotted are the PDFs of the current 10-Hz data (red) and previous DLR data (blue) for the lean mixture fraction bin ($\xi=0.075-0.125$).
Figure 6.7: Probability density functions of temperature and species mole fractions for rich mixture fraction conditions at x/D=20. Plotted are the PDFs of the current 10-Hz data (red) and previous DLR data (blue) for the rich mixture fraction bin (ζ=0.475-0.525).
Figure 6.8: Probability density functions of temperature and species mole fractions for near stoichiometric mixture fraction conditions at x/D=40. Plotted are the PDFs of the current 10-Hz data (red) and previous DLR data (blue) for the stoichiometric mixture fraction bin (ξ=0.275-0.325).
6.2 Comparison of 10-kHz Measurements with Previous Results

Figure 6.9 shows the mean and RMS fluctuations of species mole fractions and temperature (K) as a function of radial distance (mm) for measurements in the H3 flames acquired at 10 kHz using the HEPBLS as described in Chapter 3. The solid red symbols show the mean values of the species mole fractions and temperature averaged from approximately 170 bursts, each consisting of 150 images. Each data point represents the average over a 160-µm radial distance. Measurements were acquired with average laser energies of 440 mJ per pulse. As described in Chapter 3, each 1D Raman/Rayleigh line segment is 6 mm; thus, three separate 1D imaging regions are required to describe the majority of the radial extent of the flowfield (20 mm) at x/D = 10 and 20. At an axial position of x/D = 40, the same three radial positions are measured, which does not cover the full radial extent of the flow field. For all axial locations, the radial gaps between the reported measurements correspond to the physical gaps between the radial spans of the three distinct measurement locations. Each radial location represented a different set of bursts, each of which took approximately two hours to complete, implying there is possible measurement drift during individual burst sets. The error bars, plotted only on the center radial position, correspond to $2\sigma$ from the laminar calibration flame results averaged over all equivalence ratios, which represent the precision of the measurements. The open red symbols correspond to the RMS fluctuations for the current 10-kHz scalar measurements. As in Section 6.1, the data plotted in blue (solid symbols = mean; open symbols = RMS fluctuations) corresponds to the species mole fractions and temperature data from DLR [2] and available through the TNF website. It should be noted that the
reported data at \(x/D=10\) was taken on a different day than the data reported at \(x/D=20\) and \(x/D =40\), and therefore have different calibrations. Although not necessarily immediately apparent, the effects of daily calibration can be observed in the oxygen mole fraction values. For the \(x/D=10\) measurements, an expected zero \(O_2\) value is measured as a small positive value due to the constraints of the calibration procedures described in detail in Chapter 4. However, for the \(x/D=40\) data, an expected zero \(O_2\) mole fraction conditions result in systematically lower results than for \(x/D =10\), resulting in small negative values.

Overall, the current mean and RMS values from the 10-kHz measurements shown in Figure 6.9 agree well with the previous data taken at DLR. As described in Sec 6.1 in reference to the 10-Hz data, there is some difficulty in measuring zero values of oxygen and hydrogen mole fractions under rich and lean conditions, respectively. For the \(x/D=10\) measurements, the positive baseline of oxygen mole fraction leads to a lower mixture fraction as compared to that measured at DLR (and in comparison to the current 10-kHz data described in Section 6.1). At \(x/D=20\), the mean values of \(H_2O\) are lower for the current 10-kHz measurements as compared to the previous DLR measurements, although they are within the estimated uncertainty in the measurement (see Chapter 5).

Figure 6.10 shows the scatter plots of the current 10-kHz measurements. The data reported is from 170 bursts corresponds to individual measurements separated in space by 2 mm and in time by 4 ms. This corresponds to data at approximately 3-4 radial positions per axial position. The 10-kHz results shown here are very similar to that reported previously for the 10-Hz data, though the 10-kHz measurements at \(x/D=10\) only were
taken near the jet centerline and the measurements at x/D=40 only cover approximately 1/3 of the radial extent of the flowfield. The scatter follows the adiabatic flame curve very well with the exception of some small shifting in mixture fraction space, most likely due to finite noise effects and non-zero O₂ and H₂ mole fractions under rich and lean conditions, respectively. At x/D=10, the positive oxygen baseline shifts the deduced mixture fraction towards ξ=0. The negative oxygen values measured at x/D=40 shift the measurements towards ξ=1. One important note is the fact that only the measurements at x/D = 20 contain samples from the full radial extent of the flow field (see Figure 6.9); the measurements at x/D = 10 and 40 contain measurements only near centerline. In this manner, it is expected that the x/D = 20 measurements yield the most accurate representation of the quality of the 10-kHz measurements in terms of comparison with previous DLR results. Furthermore, it is plausible that the scatter plots shown in Figure 6.10 at axial positions of x/D = 10 and x/D=40 may be slightly biased towards richer samples due to the limited measurement locations. Also, measurements of x/D=40 for ξ>ξₕ are unreliable due to the small negative values of O₂ that lead to physically-meaningless values under these conditions.
Figure 6.9: Radial profiles of the mean and RMS fluctuations of major species mole fraction and temperature in the H3 flame. Current measurements from the OSU high-speed Raman system at 10 kHz are plotted in red and measurements taken at DLR are plotted in blue. The solid markers represent the mean, while the open symbols represent the RMS fluctuations.
Figure 6.10: Scatterplots of the major species mole fractions and temperature in mixture fraction space for the H3 flame. Measurements are shown for x/D = 10, 20, 40. Current measurements from the high-speed Raman system at 10 kHz are plotted as red symbols and the adiabatic flame conditions plotted as black lines.
Figure 6.11 shows the conditional averages of temperature and species mole fractions as a function of mixture fraction. All measurements at a given axial position were combined and separated into 0.05-wide mixture fraction bins. Each bin with at least 0.5% of the total number of measurement samples was averaged and plotted as a function of the mean mixture fraction for a given bin. As before, the red symbols correspond to the current data acquired at 10 kHz, while the blue symbols correspond to the previously-published data from DLR. Also shown in Figure 6.11 are the adiabatic, equilibrium flame solutions for each scalar, plotted as a solid, black line.

Since the 10-kHz measurements do not extend as far radially as the DLR data (or the current 10-Hz measurements) at x/D=10, the conditional averages at x/D = 10 do not cover the entire mixture fraction domain and are limited to $\xi > 0.5$. As before, this may bias the results and the x/D = 20 measurements are considered as the best representation of a comparison between the current 10-kHz measurements and the previously-reported DLR measurements.

As shown in Figure 6.9, a positive baseline of oxygen is seen at x/D=10. The positive values of oxygen mole fraction skew the mixture fraction towards lower values which leads to an effective shift in mixture fraction space between the current 10-kHz measurements and the DLR data. At x/D=20, the current 10-kHz data matches the DLR data very well, although there appears to be small negative oxygen mole fraction values near the stoichiometric mixture fraction. This negative oxygen value seems to cause a slight departure from the adiabatic flame curve, which is best seen in the water measurements. At x/D=40, the conditionally-averaged data matches the DLR data and the
adiabatic flame curves very well for $\xi < \xi_s$, but there are significant deviations from the adiabatic flame curve for mixture fraction values of $\xi > \xi_s$. As the $x/D = 40$ data is beyond the flame tip, these deviations from the adiabatic flame curve are an artifact of binning together data sets that contain small negative oxygen values. This facet is not observed in the DLR data, presumably due to the fact that negative species values are filtered out as part of the data processing. It is further noted that while the conditional values at $x/D=40$ are inaccurate for $\xi>\xi_s$, the mean values in physical space (Figure 6.9) were accurate.
Figure 6.11: Conditionally-averaged values of major species mole fraction and temperature in the H3 flame. Measurements are shown for x/D = 10, 20, 40. Current measurements from the high-speed Raman system at 10 kHz are plotted in red and measurements taken at DLR plotted in blue. The single-shot results are conditionally averaged within distinct mixture fraction intervals of 0.5 and plotting is limited to bins with samples containing more than 0.5% of the total number of measurement samples.

The adiabatic flame conditions are shown as black lines.
Figures 6.12, 6.13, and 6.14 show the PDFs of temperature and species mole fractions for selected mixture fraction bins similar to that shown in Figures 6.5 to 6.7 for the 10-Hz data. The results for the stoichiometric bin ($\xi=.275-.325$) at x/D = 20 is shown in Figure 6.12. Consistent with the results presented for the 10-Hz data (Figure 6.5), the current 10-kHz measurements and DLR measurements have similar mean values and standard deviations. The PDFs of the lean mixture fraction bin ($\xi=.075-.125$) are shown in Figure 6.13. Overall, the agreement between the PDFs is good for this selected mixture fraction range, with only the temperature and H$_2$O PDFs showing significant broadening as compared to the DLR measurements. However, the mean values between the two data sets are nearly identical and the additional broadening likely is due only to measurement noise. Figure 6.14 shows the PDFs for a fuel-rich mixture fraction bin ($\xi=.475-.525$). While a decrease in the peak and a broadening of the PDF is expected for this mixture fraction range, the standard deviation of the PDFs for the 10-kHz data is significantly larger than that observed with the 10-Hz data and in comparison with the DLR data. This is likely due to oxygen measurements and in particular, the small negative bias in the O$_2$ values for slightly rich conditions (see Figure 6.11). As observed in the 10-Hz data, small negative offsets in the O$_2$ values near stoichiometric conditions can lead to an effective shift in mixture fraction space for the scalar values, suggesting that the accuracy of the mixture fraction measurement is skewed by small negative oxygen values and hence PDFs for a given mixture fraction range will be broadened. The PDFs of the temperature and species mole fractions for the stoichiometric bin ($\xi=.275-.325$) at x/D=40 shown in Figure 6.15 are consistent with the trends observed at 10 Hz (Figure 6.8). While the shift
in the temperature and H₂O PDFs are in the opposite direction as compared to the 10-Hz data, shifts towards lower temperatures and H₂O mole fractions are expected due to the small negative values of oxygen and hydrogen.

This chapter has presented detailed comparisons between the current data using the OSU high-speed Raman/Rayleigh imaging system and previously-published data from DLR in the H3 flame. Focusing solely on the high-speed measurements for the moment, very good agreement was observed for the mean and RMS fluctuations for temperature and species in physical space; that is, as a function of radial position (Figure 6.9). Small negative offsets in O₂ mole fraction lead to small discrepancies in the computed mixture fraction (ζ), which in turn leads to small, but observable differences in statistics conditioned on mixture fraction. However, the remainder of the dissertation focuses on using the newly developed high-speed Raman/Rayleigh imaging capabilities to determine time-dependent characteristics of turbulent nonpremixed flames, including correlation functions and time scales as function of spatial position and not conditioned on mixture fraction. In this manner, the single-shot measurements are of sufficient quality and precision to investigate the spatio-characteristics of the scalar fields within the flame.
Figure 6.12: Probability density functions of temperature and species mole fractions near stoichiometric conditions at x/D=20. Plotted are the PDFs of the current 10-kHz data (red) and previous DLR data (blue) for the stoichiometric mixture fraction bin ($\xi=0.275-0.325$).
Figure 6.13: Probability density functions of temperature and species mole fractions for lean mixture fraction conditions at x/D=20. Plotted are the PDFs of the current 10-kHz data (red) and previous DLR data (blue) for the lean mixture fraction bin (ξ=0.075-0.125)
Figure 6.14: Probability density functions of temperature and species mole fractions for rich mixture fraction conditions at x/D=20. Plotted are the PDFs of the current 10-kHz data (red) and previous DLR data (blue) for the rich mixture fraction bin ($\xi=0.475-0.525$).
Figure 6.15: Probability density functions of temperature and species mole fractions for near stoichiometric mixture fraction conditions at x/D=40. Plotted are the PDFs of the current 10-kHz data (red) and previous DLR data (blue) for the stoichiometric mixture fraction bin ($\xi=0.275-0.325$).
Chapter 7: Temporally-resolved Measurements and Analysis

In the previous chapter, 10-kHz measurements acquired with the high-speed 1D Raman/Rayleigh scattering imaging system at Ohio State were assessed and validated through statistical comparisons with previously-published 10-Hz measurements available for the H3 (Re=10,000) flame from DLR. This chapter will extend analysis beyond low-order statistical descriptions (e.g., mean and RMS) and focus on extracting new temporal information contained within the measurements. First, time-resolved visualizations are presented in order to provide a qualitative description of the coupled spatio-temporal dynamics contained within the turbulent flames. Next, temporally-based statistics, including temporal autocorrelations and the associated integral time scales of all available scalars (N\textsubscript{2}, H\textsubscript{2}, O\textsubscript{2}, H\textsubscript{2}O, mixture fraction, and temperature) will be presented, in order to describe the relevant time scales of the flow and more specifically, to illuminate the differences between specific scalars and describe the turbulence-chemistry interaction. Finally, the temporal cross-correlations and timescales of important scalar pairs will be discussed.
7.1 High-Speed Visualization

7.1.1 Sample Raman Images at 10 kHz

Figure 7.1 shows a sample set of 10 sequential time-resolved Raman scattering images acquired at 10 kHz. The sample data is extracted from a longer 150-image sequence. The 1D line segments (y axis) cover 6 mm, centered at a radial position of 4 mm from the centerline. The dispersed Raman scattering (x-axis) is collected over wavelengths corresponding to 559-704 nm (left to right). The images shown in Figure 7.1 were acquired at x/D=20, where centerline values consist mostly of low-temperature fuel (50% H₂/50% N₂) mixing with combustion products (N₂ and H₂O). It is important to note that the raw signals are directly proportional to number density, and therefore are influenced not only by species mole fraction, but temperature as well. This causes water, a high-temperature combustion product, to appear faint as compared to the relatively cold fuel species (H₂ and N₂). As an example of the observed dynamics, the far right hydrogen line is shown to exit and then return to the field of view. This corresponds to a relative decrease in the N₂ Raman signal and an increase in the H₂O Raman signal; all of which suggest the occurrence of combustion products either from mixing or the sudden passing of an eddy of combustion products through the field of view.

Figure 7.2 shows another sample ten-image sequence acquired at 10 kHz. While still at an axial position of x/D=20, the field of view in this image sequence was centered at a radial position of 12 mm from centerline, which is near the average position of the stoichiometric contour as shown in Chapter 6. At this radial position, the majority of the fluid corresponds to burnt combustion products with intermittent bursts of unburned fuel
and cold air entering the field of view. At Δt=0, cold air is seen entering the field-of-view as determined by the simultaneous occurrence of high oxygen and nitrogen signals. At Δt = 100 μs, the oxygen signal and the nitrogen signal increase, meaning a continual increase in the number density of air, which could correspond to locally lower temperatures, higher mole fractions, or both. This is consistent with entrainment from the coflow into the field of view. From Δt = 100-500 μs, the oxygen signal decreases, suggesting either the air was transported out of the field-of-view or was consumed by the flame via reaction. Starting at Δt = 500 μs, unburned gases (N₂ and H₂) enter the field-of-view and then the signal disappears either through transport or reaction. In any manner, the raw images give a qualitative insight on the local dynamics of the flow field and the intermittent nature of the local species number density at a specific spatial location.

Figures 7.3 to 7.6 compare sample single-shot images extracted from the 10-kHz image sequences with the mean image in order to highlight the fluctuating nature of the flow. While some instantaneous images (such as that in Figure 7.6) look very similar to the mean image, others sample images (such as those in Figures 7.4 and 7.5) show significant differences between the instantaneous and mean images, giving insight on the degree of species fluctuation at a given position. As one example, the instantaneous image in Figure 7.3 is quite interesting as the boundary between the hot and cold flows is observed by the occurrence of the rotational hydrogen lines, which are not present in cold fuel.
Figure 7.1: Sample 10-kHz Raman scattering images acquired using the high-speed 1D Raman/Rayleigh scattering system at Ohio State. These ten sample images were taken at x/D=20, r=1-7 mm (bottom to top) and are separated in time by 100 µs. Spectral lines for O$_2$ (580 nm), N$_2$ (607 nm), H$_2$O (661 nm), H$_2$ (683 nm), and H$_2$,rot (560-580 nm).
Figure 7.2: Sample 10-kHz Raman scattering images acquired using the high-speed 1D Raman/Rayleigh scattering system at Ohio State. These ten sample images were taken at $x/D=20$, $r=9-15$ mm (bottom to top) and are separated in time by 100 µs. Spectral lines for $O_2$ (580 nm), $N_2$ (607 nm), $H_2O$ (661 nm), $H_2$ (683 nm), and $H_2_{rot}$ (560-580 nm).
Figure 7.3: Raman scattering images acquired at an axial position of x/D=10 and centered at a radial position of 3 mm from centerline using the high-speed 1D Raman/Rayleigh scattering system at Ohio State. (Top) Average image from 175 bursts of 150 10-kHz images. (Bottom) An instantaneous image extracted from a 10-kHz image sequence.
Figure 7.4: Raman scattering images acquired at an axial position of x/D=20 and centered at a radial position of 4 mm from centerline using the high-speed 1D Raman/Rayleigh scattering system at Ohio State. (Top) Average image from 175 bursts of 150 10-kHz images. (Bottom) An instantaneous image extracted from a 10-kHz image sequence.
Figure 7.5: Raman scattering images acquired at an axial position of $x/D=20$ and centered at a radial position of 13.3 mm from centerline using the high-speed 1D Raman/Rayleigh scattering system at Ohio State. (Top) Average image from 175 bursts of 150 10-kHz images. (Bottom) An instantaneous image extracted from a 10-kHz image sequence.
Figure 7.6: Raman scattering images acquired at an axial position of x/D=40 and centered at a radial position of 2.5 mm from centerline using the high-speed 1D Raman/Rayleigh scattering system at Ohio State. (Top) Average image from 175 bursts of 150 10-kHz images. (Bottom) An instantaneous image extracted from a 10-kHz image sequence.

While raw images can give a qualitative idea of the physical phenomena occurring within the flow, the processed Raman/Rayleigh data allows examining the major species, temperature, and mixture fraction simultaneously as a function of time and space. Shown in Figure 7.7 is a 2D spatio-temporal map of the processed data from a single burst at x/D of 20 and centered at 4 mm from the jet centerline. The vertical axis
represents the spatial position (radial distance from centerline), the horizontal axis represents the time within a single burst, and the color map represents the scalar value. Using this type of spatio-temporal map, the local dynamics, including the spatial and temporal intermittency of the scalar fields can be observed at a given axial position. As an example, from 6-9 ms, the emergence of the flame front is seen near centerline. This is determined by the local increase in temperature and water mole fraction and the simultaneous decrease in oxygen and hydrogen mole fraction. Mixture fraction also slightly decreases, suggesting the influx of flame products mixed with entrained air. An interesting feature occurs around 13 ms, where a small amount of “colder” (T ~ 1200 K) air is seen entering the field of view, which corresponds to the negligible hydrogen levels, a “break” in the water structure, and low mixture fraction.

Another spatio-temporal map, centered at x/D=20 and r=12 mm is shown in Figure 7.8. At this axial position, the average position of the stoichiometric contour is located at 11 mm from the centerline and is contained within the radial domain shown in Figure 7.8. As expected, the flow fluctuates on slower timescales at this radial position due to decreased velocities and lower local Reynolds numbers (increase in local viscosity due to higher temperatures). This is confirmed by the observed slower transitions from hot-to-cold temperatures and high-to-low scalar measurements and the increase in size of the flow features. At t = 12 ms, a burst of hot combustion products is seen entering the probe volume followed by cooler air entrained from the coflow. Both processes occur over 1-2 ms, demonstrating the longer time scales at the present radial location.
Figures 7.9 and 7.10 each provide a time trace for a single spatial position along the laser line during a single burst. At x/d = 20 and r = 1.5 mm from the centerline (Figure 7.9) there are high-frequency fluctuations in the scalar values which results in large temporal gradients; however, the actual magnitude of the fluctuations are not that large as most of the gases in the flow originates from the fuel jet, though the occasional observance of burnt gases (t=0 ms) is seen. As the radial position is increased to r =12 mm (Figure 7.10), which is near the average position of the stoichiometric contour, there is more interaction between fuel, coflow air, and burned combustion products. The time trace shown in Figure 7.10 shows evidence of mixing between combustion products and the coflowing air (0-3 ms), a possible occurrence of the high-temperature reaction zone (~7 ms), and mixing between fuel and hot gases (~12 ms).
Figure 7.7: 2D Spatio-temporal map of scalar data acquired at x/D=20 and centered at a radial position of 2.5 mm. Results are from a single 10-kHz. Each vertical strip represents the binned “superpixels” along the 1D laser line for a given scalar in physical space. The strips are stacked horizontally (left to right) in the order of time they were acquired (100 µs spacing). The result is a 2D map of the scalar with the colors signifying temperature, mole fraction, or mixture fraction.
Figure 7.8: 2D Spatio-temporal map of scalar data acquired at x/D=20 and centered at a radial position of 12 mm. Results are from a single 10-kHz. Each vertical strip represents the binned “superpixels” along the 1D laser line for a given scalar in physical space. The strips are stacked horizontally (left to right) in the order of time they were acquired (100 µs spacing). The result is a 2D map of the scalar with the colors signifying temperature, mole fraction, or mixture fraction.
Figure 7.9: Single 10-kHz time trace of temperature and species mole fraction acquired at an axial position of $x/D=20$ and a radial position of 1.5 mm from the centerline.
Figure 7.10: Single 10-kHz time trace of temperature and species mole fraction acquired at an axial position of x/D=20 and a radial position of 12 mm from the centerline.

Figure 7.11 shows the temporal fluctuation of the measured scalars within a single 15-ms burst as a function of mixture fraction, which describes the state of mixing. The results are from a single superpixel at an axial position of x/D =20 and a radial position of...
12 mm from centerline. Also shown in Figure 7.11 are the average values of the scalars (from all bursts) conditioned on mixture fraction, plotted as solid lines. It is seen that the measurements from a single 15-ms burst cover a wide range of mixture fraction values, highlighting the intermittency of the system. It is interesting to note that for samples corresponding to $\xi < \xi_s$, the scalars lie close to the conditional mean, but for scalar values corresponding to $\xi > \xi_s$, there is more deviation from the conditional average. The radial position of 12 mm from centerline corresponds to an average mixture fraction value of $\xi < \xi_s$, thus it is possible that the instantaneous scalar values that lead to $\xi > \xi_s$ are the result of highly intermittent bursts from the centerline which lead to large, instantaneous departures from the conditional average. In addition, a significant portion of the scatter may be attributed to uncertainties in the mixture fraction (for $\xi > \xi_s$) due to difficulties in measuring zero oxygen mole fraction as discussed in Chapter 6.
Figure 7.11: 10-kHz scalar fluctuations (in mixture fraction space) within a single 15-ms burst at an axial position of $x/D=20$ and a radial position of $r=12$ mm from the centerline. The stoichiometric mixture fraction is plotted as a dashed vertical line and solid lines represent the average values conditional on mixture fraction at $x/D=20$. 
7.2 Scalar Autocorrelations and Time Scales

While the visualizations in Section 7.1 offer qualitative insight into the local mixing dynamics and turbulence-chemistry interaction, flow statistics offer a way quantify the behavior of the turbulent combustion processes; and in particular, the true utility of the current measurements lies in the fact that new, temporally-based information is available for the first time. This section will focus on the determination of temporal autocorrelation functions for temperature and major species; the latter of which has not been previously available within the literature. The importance of the autocorrelation function is that it yields a representation of the “memory” of the process or more specifically, the time over which one quantity is correlated with itself. From the auto-correlation functions, the integral time scales will be derived in order to give new insight into how the largest turbulent time scales vary amongst different scalars within a turbulent combustion environment.

In order to reduce the effects of noise, the reported autocorrelation functions are correlation functions between neighboring radial superpixels (each of size 160 μm) such that:

$$\rho(t') = \frac{<\phi'_1(t)\phi'_2(t')>}{\sigma_1\sigma_2}$$  \hspace{1cm} \text{Equation 7.1}

where $t'$ represents a time lag with respect to time $t$, $\phi'_1$ and $\phi'_2$ are the scalar fluctuations at pixels 1 and 2, respectively and $\sigma_1$ and $\sigma_2$ are the standard deviations of the measured scalar fluctuations at pixels 1 and 2, respectively. As shown in [14], this approach has the benefit of averaging out uncorrelated noise of the detector and
recovering the true auto-correlation function over a spatial volume of 320 μm.

Autocorrelation functions for each scalar were calculated from approximately 175 bursts at each radial position using the Matlab function ‘xcorr’ and normalized using the average and RMS calculated at each position from the 175 bursts. While this process was repeated at every radial position, only a limited number of results at selected spatial positions are reported here.
Figure 7.12: Average autocorrelation functions of temperature, major species, and mixture fraction at x/D=20.

(Continued)
Figures 7.12 and 7.13 show the average autocorrelation functions for temperature, major species, and mixture fraction at an axial position of x/D=20. Figure 7.12 shows the auto-correlation functions for all measured scalars at particular radial locations, while Figure 7.13 shows the calculated auto-correlation functions for each individual scalar as a function of radial position. Figures 7.12 and 7.13 indicate that for all scalars, the decorrelation times (area under the auto-correlation function) increases with increasing
radial position. As shown in Figure 7.12, near the centerline (r=1 mm) and the co-flowing air stream (e.g., r = 17 mm and 20 mm), the de-correlation times for all measured scalars are similar. However, for radial positions near the average position of the stoichiometric contour (e.g., r = 6 mm and 11 mm), the calculated auto-correlation functions vary significantly across the scalars. These radial positions correspond to the regions with the majority of the chemical reaction and heat release (on average), which appears to lead to variations in the de-correlation times amongst the various scalars. As one example, at r = 6 mm, the area under the auto-correlation functions increases by approximately a factor of three over all scalars with temperature and water showing the fastest de-correlation, followed by hydrogen and mixture fraction, which displays the slowest de-correlation. At r = 11 mm, the differences in the de-correlation times across all scalars decrease to approximately a factor of two, where temperature and water still display the fastest de-correlation times, followed by oxygen and mixture fraction. For the radial positions near the average position of the stoichiometric contour, the auto-correlation function of the mixture fraction is closely tied to the reactant species (H\textsubscript{2} in fuel rich conditions and O\textsubscript{2} in fuel lean conditions) and quite different than that of temperature.
Figure 7.13: Average autocorrelation functions for temperature, major species, and mixture fraction at various radial positions and an axial position of $x/D=20$. 
In order to quantify the timescales associated with each scalar, the integral time scales of each scalar was calculated and shown in Figure 7.14 as a function of radial position. The integral time scale, \( \tau \), typically is defined as:

\[
\tau = \int_0^\infty \rho(t') dt' \quad \text{Equation 7.2}
\]

However, in practice a time series of infinite duration is not available and the measured auto-correlation functions must be evaluated over finite time durations. Furthermore, for long time lags, the uncertainty in the tail of the auto-correlation function increases due to an increasingly fewer number of samples as compared to shorter time lags. In this work, the area under each measured autocorrelation function was evaluated from \( t'=0 \) to the time lag where the autocorrelation function decayed to a value equal to \( 1/e^2 \), denoted \( t_{e2} \). Furthermore, if the autocorrelation function is assumed to take the shape of exponential decay, which is the functional form under high-Reynolds number conditions, the area under the autocorrelation function from \( t' = 0 \) to \( t' = t_{e2} \) is:

\[
\int_0^{t_{e2}} \rho(t') = (1 - \frac{1}{e^2})\tau \quad \text{Equation 7.3}
\]

In this manner, the integral time scales for the scalars estimated using:

\[
\tau = \left( \frac{1}{1 - e^2} \right) \int_0^{t_{e2}} \rho(t') \quad \text{Equation 7.4}
\]

In order to ensure all reported integral time scales were calculated from data which was not overly influenced by measurement noise, a SNR threshold was applied to the data, which was established from the laminar calibration flame data presented in Chapter 5. Specifically, temperature and mole fraction values that had a SNR, defined as the scalar mean divided by the scalar RMS in the near-adiabatic Hencken flames, greater than a
certain threshold were used to compute the auto-correlation functions and corresponding integral time scales. As one specific example, this approach was implemented to avoid calculating \( \tau \) for quantities such as \( \text{H}_2 \) or \( \text{O}_2 \) near the stoichiometric contour, where the mole fraction values would be near zero due to chemical reaction. In the current work, the SNR threshold was chosen to be 2, although there was little change in the results when the parameter was varied between 1 and 4.

In general, Figure 7.14 shows that the derived integral time scales for the species, temperature, and mixture fraction increase with increasing radial position, which is consistent with the qualitative observations from the auto-correlation functions shown in Figure 7.12 and Figure 7.13. However, the integral time scales of the fuel and oxidizing species (\( \text{H}_2 \) and \( \text{O}_2 \)) appear to be an exception to this general observation near the stoichiometric contour due to chemical reaction. It is expected that for any passive scalar in the jet flame, the integral time scales would increase with increasing radial distance since the local velocities decrease and the local temperature and viscosity increase yielding larger characteristic length scales. This is consistent with the observation for the integral time scales of the mixture fraction, which is a conserved scalar; however, the rapid consumption of \( \text{H}_2 \) and \( \text{O}_2 \) in the regions near the stoichiometric contour leads to shorter integral time scales than would be expected for a non-reacting scalar. Near centerline, the measured integral time scales are comparable, but as radial position increases towards the stoichiometric contour, a large spread in the integral time scales amongst the scalars is observed, where the integral time scales for hydrogen and mixture fraction are much larger than that of temperature and water. Near the stoichiometric
contour ($r = 11$ mm), $\tau_\xi \sim 3\tau_T$ and is significantly larger than both $\tau_{H_2O}$ and $\tau_{H_2}$. For increasing radial positions ($r > 11$ mm; $\xi < \xi_s$), $\tau_\xi$ closely follows $\tau_{O_2}$. Overall, it is apparent that all of the scalars within the nonpremixed flame system cannot be described by a single characteristic (integral) time scale, at least for spatial positions near the stoichiometric contour, where the majority of the chemical reaction takes place.

Figure 7.14: Calculated integral time scales ($\tau$) at $x/D=20$ as a function of radial position. The ‘dashed line’ shows the average position of the stoichiometric contour, $\xi_s$. 
Figure 7.15 shows the autocorrelation functions for radial positions of 0 and 20 mm from centerline at an axial position of x/D=40 calculated with the same process as described above. As expected, the integral time scales increase for an increasing axial position. Because x/D=40 is just above the average position of the flame tip, chemical effects on the measured auto-correlation functions are still observed, although much less than the observed effects at x/D = 20. Specifically, the de-correlation times appear to vary amongst the scalars near centerline to a small degree. At r = 0, the de-correlation of the mixture fraction closely follows that of oxygen while the auto-correlation functions of hydrogen, water, and temperature are very similar. Further out radially at r = 20 mm from centerline, all the autocorrelation functions fall on top of each other, implying that the flow can be described by a pure-mixing regime. For comparison, the autocorrelation functions of the various scalars are plotted at multiple radial positions in Figure 7.16 and multiple axial positions in Figure 7.17 and Figure 7.18. As expected, decorrelation times (characteristic time scale) increases with both radial and axial position.
Figure 7.15: Average autocorrelation functions of temperature, major species, and mixture fraction at x/D=40.
Figure 7.16: Average autocorrelation functions for temperature, major species, and mixture fraction at various radial positions and an axial position of at x/D=40.
Figure 7.17: Average autocorrelation functions for temperature, major species, and mixture fraction at a radial position of 1 mm from the centerline. Results are shown for two axial positions corresponding to $x/D = 20$ and 40.
Figure 7.18: Average autocorrelation functions for temperature, major species, and mixture fraction at a radial position of 11 mm from the centerline. Results are shown for two axial positions corresponding to $x/D = 20$ and 40.

More quantitatively, the calculated integral time scales are shown in Figure 7.19 for an axial position of $x/D = 40$. Similar to the results observed at $x/D=20$ for radial positions larger than that corresponding to the average position of the stoichiometric mixture fraction, the integral time scales of mixture fraction and are very similar. As a general observation at $x/D = 40$, as the radial position increases away from centerline it appears that hot combustion products mix with coflowing air in what resembles a pure mixing problem. For this situation, there is little to no effect of chemistry on the derived...
integral time scales. In this regime, the flow would be well described by a pure mixing problem and adequately described by a single characteristic integral time scale.

Figure 7.19: Calculated integral time scales (τ) at x/D=40 as a function of radial position.
7.3 Scalar Cross-correlations and Timescales

This section presents a new type of analysis made available from the time-resolved, multi-scalar measurements, namely the determination of temporal cross-correlations and the subsequent determination of the associated time scales between various scalar pairs. Similar to the auto-correlation function (for a single scalar) defined by Equation 7.1 in Section 7.2, the temporal cross-correlation between two scalars is defined as:

\[ c_{12}(t') = \frac{\langle \varphi'_1(t) \varphi'_2(t') \rangle}{\sigma_1 \sigma_2} \]  

Equation 7.5

where \( \varphi'_1 \) and \( \varphi'_2 \) are the fluctuations of different scalars 1 and 2 and \( \sigma_1 \) and \( \sigma_2 \) are the standard deviations of the measured scalar fluctuations for different scalars 1 and 2, respectively. The scalar cross-correlations yield a representation of how (and on what timescales) various scalar pairs interact within the turbulent flame. Similar to the calculations of the autocorrelation functions, the temporal cross-correlation between scalars was calculated from approximately 175 bursts at each radial position. The cross-correlations reported in this section utilized the same radial superpixel position on two different scalar channels. Since the cross-correlation is determined from two separate scalar channels (two distinct pixel locations within the detector), the effects of the uncorrelated noise are mitigated in the same manner as using two adjacent “radial superpixels” for the determination of the auto-correlation functions. The temporal cross-correlation functions are determined using the Matlab function ‘xcorr’ and normalized using the average and RMS fluctuation calculated for each scalar at each position from the 175 bursts.
Figure 7.20 shows the cross-correlation function between hydrogen mole fraction and the other measured flow field scalars. The correlation of hydrogen with mixture fraction at $t' = 0$ is near unity for all reported radial positions ($r = 0, 6,$ and $11$mm from centerline), demonstrating the strong, positive correlation between hydrogen and mixture fraction. Similar to the autocorrelation functions reported in Section 7.2 for a single scalar, as the radial distance increases from centerline, the de-correlation time or alternatively, the “interaction time” between the two scalars increases. As expected, the correlation of hydrogen with oxygen is negative at the single reported radial position (near the stoichiometric contour) and hydrogen is strongly negatively correlated with water near centerline. Since water is a combustion product, on average hydrogen is consumed in order to produce water. However, near the stoichiometric contour, there is a very weak positive correlation between hydrogen and water. This likely indicates that, for radial positions, which on average correspond to $\xi < \xi_s$, any hydrogen (which has diffused past $\xi_s$) mixes with combustion products with little chemical reaction. A similar relationship between hydrogen and temperature is observed as that for hydrogen and water.

Figure 7.21 shows the derived cross-correlation functions for oxygen mole fraction with the remaining measured scalars at $x/D=20$. As expected, negative correlations exist for all scalars with oxygen, although the degree of the anti-correlation and the associated time scales vary amongst the scalar pairs and the exact radial position.
Figure 7.20: Average cross-correlation functions for hydrogen mole fraction and remaining scalars at various radial positions and an axial position of $x/D=20$. 
Figure 7.21: Average cross-correlation functions for oxygen mole fraction and remaining scalars at various radial positions and an axial position of x/D=20.

Figure 7.22 shows the cross-correlation functions between water and the remaining measured scalars. Water and mixture fraction appear to have a relationship that is highly dependent on spatial position. In fuel rich parts of the flame (r = 1 mm), water and mixture fraction have a strong negative correlation as the formation of water implies chemical conversion of hydrogen + oxidizer to products, which decreases the local mixture fraction. However, as the radial position increases towards the position...
corresponding to the stoichiometric contour \((r = 11 \text{ mm})\), a strong, positive correlation between the mixture fraction and water is observed. This can be explained by considering a typical state relationship between water and mixture fraction. For fast chemistry, the state relationship between the two species consists of a piecewise linear relationship, where \(dX_{H_2O}/d\xi\) is negative for fuel rich conditions and \(dX_{H_2O}/d\xi\) is positive for fuel lean conditions. At stoichiometric conditions, \(dX_{H_2O}/d\xi \sim 0\), which would imply that any small fluctuation in the local mixture fraction would give rise to a similar small fluctuation in water, thus positive correlation. As expected, the relationship between temperature and the remaining scalars (Figure 7.23) is nearly identical to that observed between water and the remaining scalars (Figure 7.22). Based on the results reported in Chapter 6 which showed that the mean and RMS profiles of temperature and water display the same functional form over all physical space and mixture fraction state space, the cross-correlation of temperature and water (Figure 7.22) closely resembles the autocorrelation of either species as reported in Section 7.2. Furthermore, correlation between temperature and other measured scalars (Figure 7.22) should display the same functional relationship as that of water and the remaining scalars.
Figure 7.22: Average cross-correlation functions for water mole fraction and remaining scalars at various radial positions and an axial position of x/D=20.
Figure 7.23: Average cross-correlation functions for temperature and remaining scalars at various radial positions and an axial position of x/D=20.

The scalar-interaction timescales (taken from the cross-correlations) were calculated in a similar manner as the integral time scales described by in Section 7.2. Due to the nature of the cross-correlations, additional SNR threshold criteria were added to ensure that the reported time scales were not overly corrupted by measurement noise.

First, both scalar values (for a given scalar-scalar pair) considered in the cross-correlation needed to have a SNR, defined as the scalar mean divided by the scalar RMS in the near-adiabatic Hencken flames, of greater than two. Second, the value of the cross-correlation
at time $t'=0$ needed to be greater than 0.2. Results from these calculations at $x/D=20$ are shown in Figure 7.24 as a function of radial position, along with the previously reported integral timescales for the two individual scalars.

Since the derived scalar-interaction timescale represents a measure of an interaction time between two scalars, it is logical to assume that the scalar-interaction timescale should not exceed the time at which either of the two scalars is correlated with itself, i.e., the integral time scale. In general, it is expected that the scalar-interaction timescales would be bound by the individual scalar integral time scales. While this trend is observed for the measurable scalar pairs near centerline and near the co-flowing air stream, this is not observed at radial positions approaching the stoichiometric mixture fraction position (i.e., region of high chemical reaction). At this time, it is not known whether this is a direct indication of the effects of chemical reaction/heat release or an artifact of the data processing, i.e., the difficulty in measuring small oxygen and hydrogen mole fractions near the stoichiometric contour.
Figure 7.24: Cross-scalar time scales as a function of radial position at x/D=20.

(Continued)
Figures 7.25 to 7.27 plot the scalar cross-correlation functions at x/D=40, an axial position located beyond the average position of the flame tip. As described in Section 7.2, this spatial location more closely resembles that of a pure mixing environment with little chemical effects. The results in Figures 7.25 to 7.27 are consistent with the auto-correlation functions shown in Section 7.2, which were very similar for all scalars. In general, the area under the cross-correlation functions (i.e., the characteristic time scale) appears to increase with both radial and axial position. Figure 7.28 shows the derived scalar scalar-interaction timescales at x/D = 40, along with the previously-reported
integral time scales for the two scalars under consideration for a given scalar-interaction timescale. As expected, the scalar scalar-interaction timescales are very similar to the individual integral time scales, further emphasizing the lack of chemical effects on the derived characteristic turbulence time scales at x/D=40.

Figure 7.25: Average cross-correlation functions for oxygen mole fraction and remaining scalars at various radial positions and an axial position of x/D=40.
Figure 7.26: Average cross-correlation functions for water mole fraction and remaining scalars at various radial positions and an axial position of x/D=40.
Figure 7.27: Average cross-correlation functions for temperature and remaining scalars at various radial positions and an axial position of x/D=40.
Figure 7.28: Scalar-interaction timescales as a function of radial position at x/D=40.
Chapter 8: Summary, Conclusions, and Suggested Future Work

The primary contribution of this research was the development of a combined high-speed 1D Raman/Rayleigh scattering laser diagnostic technique for quantitative time-resolved measurements of temperature, major species concentrations, and mixture fraction in turbulent nonpremixed flames. While combined 1D Raman/Rayleigh scattering imaging is a staple of turbulent combustion diagnostics, traditionally, it has been limited to measurements at low acquisition rates. This limits the characterization of turbulence-chemistry interaction to low-order statistical descriptions and provides no information concerning the dynamic nature of turbulent combustion environments. Alternatively, the majority of existing high-speed measurements provide qualitative visualization of the flow structure and topology, but limited quantitative metrics that can be used for the development of predictive turbulent combustion models. Specific exceptions to this include a few temperature and minor species measurements. The successful development of the high-speed 1D Raman/Rayleigh imaging approach presented here provides a significant step towards further understanding of turbulence-combustion interaction through direct visualization of turbulent flame dynamics and previously-unavailable joint space-time statistics of temperature, major species concentrations, and mixture fraction.

The high-speed 1D Raman/Rayleigh scattering imaging was facilitated through the development of a custom-built spectrometer utilizing two high-speed CMOS cameras
and a single high-speed image intensifier, the implementation of a robust data reduction methodology specifically developed for high-speed data collection, and the use of the unique High-Energy Pulse Burst Laser System (HEPBS) at Ohio State for ultra-high pulse energy output at multi-kHz repetition rates. Detailed calibrations in near-adiabatic, laminar flames were used to assess the accuracy and precision of the high-speed Raman/Rayleigh measurements. In general, the measurements in the near-adiabatic flames agreed very well with expected adiabatic flame calculations over a broad range of temperatures and equivalence ratio. One of the limitations identified with the current approach is the difficulty in measuring small concentrations of species, and in particular, small concentrations of O\textsubscript{2} near stoichiometric conditions. Measurements of small negative values of O\textsubscript{2} were found to influence equivalence ratio calculations under rich conditions. The somewhat inaccurate measurement of O\textsubscript{2} under rich conditions was due to low signal-to-noise ratios at these positions. The low SNR was a function of high flame temperatures (low signal levels due to low number density), relatively-high noise levels and limited dynamic range of the high-speed detectors, and large amounts of crosstalk in flame conditions. The non-physical O\textsubscript{2} values lead to lower signal-to-noise ratios under rich flame conditions as compared to those measured in lean conditions. While the high-speed measurements had lower SNR values than previously reported at low repetition rates, the SNR reduction was consistent with that expected with high-speed intensified CMOS cameras (as compared to low-noise, scientific-grade CCD cameras) and was sufficient for accurate time-resolved measurements of temperature and the major
species, which allows the deduction of the mixture fraction. However, it was concluded that the SNR was not high enough to accurately determine scalar gradients.

Following system characterization in the near-adiabatic, laminar flames, detailed comparisons between the current temperature and species concentration measurements from the DLR H3 turbulent jet flame using the high-speed Raman/Rayleigh imaging system and previously-reported data was performed to assess the accuracy of single-shot measurement under turbulent flame conditions. Overall the radial profiles of mean and RMS fluctuations of temperature, major species mole fractions (H$_2$, O$_2$, N$_2$, and H$_2$O), and mixture fraction agreed very well with the previously-reported data from DLR. Small negative offsets in O$_2$ mole fraction, similar to that observed in the laminar calibration flames, led to small discrepancies in the computed mixture fraction ($\bar{\xi}$) and observable differences in statistics conditioned on mixture fraction.

Following statistical assessment and validation in turbulent flames, extensive 10-kHz measurements were performed in the same H3 nonpremixed jet flame to visualize the coupled spatio-temporal dynamics and determine new temporal statistics including temporal autocorrelation functions and the associated integral times scales of all available scalars (H$_2$, O$_2$, H$_2$O, mixture fraction, and temperature). In general, the integral timescales were found to increase with increasing radial (0 to 24 mm) and axial (x/D=20 to 40) position; however the integral timescales of the reactants (H$_2$ and O$_2$) were found to be exceptions to this general rule near the stoichiometric contour due to chemical reaction effects. While the integral timescales are similar for all scalars near the centerline and well beyond flame boundaries (portions of the flow with little-to-no
influence of chemistry), the scalars behave very differently within the flame boundaries where the majority of chemical reaction and heat release occur. Differences as large as a factor of three in the associated integral timescales were observed across all scalars. Near the stoichiometric contour, the integral timescale for the mixture fraction is up to a factor of three times larger than that of temperature and water. In addition, it was observed that for radial positions near the stoichiometric contour, the autocorrelation function and integral timescales of the mixture fraction is closely tied to the reactant species (H$_2$ in fuel rich conditions and O$_2$ in fuel lean conditions). Overall, it was concluded that the scalars within the nonpremixed flame system cannot be described by a single characteristic (integral) timescale, at least for spatial positions near the stoichiometric contour, where the majority of the chemical reaction takes place. For measurements downstream of the flame tip, the autocorrelation functions and derived integral timescales were similar but with observed differences indicating small, non-negligible chemical effects. However, it is assumed that the flow farther beyond the flame tip is well described by a pure mixing problem and adequately described by a single characteristic integral timescale.

The unique high-speed multi-scalar data provided the opportunity for a new type of analysis, namely the determination of the temporal cross-correlation amongst scalar pairs and the determination of scalar-interaction timescales. Generally, the scalar interaction times calculated from the temporal cross-correlation functions were bound by the individual scalar integral timescales near centerline and near the co-flowing air stream. However, this characteristic was not observed for radial positions approaching
the stoichiometric mixture fraction and there was no discernible trend, meaning each scalar-scalar pair yielded a unique radial dependence on the derived scalar-interaction timescale. At this time, it is not known whether this is an indication of the effects of chemical reaction or an artifact of the data processing. Since the derivation of this statistic is new, additional work is needed to understand the observed trends and significance of its spatial variation.

Future work should focus on extending the newly developed measurements to turbulent nonpremixed hydrocarbon flames in order to investigate flames with more complex flame chemistry. As the RAMSES tables and Sandia polynomials used in this dissertation were designed for use in hydrocarbon flames, no new data processing methodology is needed, only additional calibration measurements in cold flows of CO, CO$_2$, and CH$_4$, and methane-based calibration flames. Most likely, increased signal-to-noise ratios would be necessary to accurately measure CO and CO$_2$ in hydrocarbon flames. Increased SNR can be achieved in two ways: (1) by increasing the signal or (2) decreasing the measurement noise. Larger signal gains in the Raman and Rayleigh signals are possible with the use of the newly-configured HEPBLS. In the data presented in this dissertation, there were large fluctuations in output laser energy due to a systematic failure in the HEPBLS master oscillator. While the effects of the energy modulation were mostly corrected, the overall laser energy used to acquire data was reduced because the camera settings had to account for the highest fluctuations in laser energy. In addition, the output beam quality of the HEPBLS was reduced due to the failure of the master oscillator, which prohibited the use of an optical pulse stretcher and
higher pulse energies. In the newest version of the HEBLs, a new seed laser has been implemented that allows for variable pulse lengths, thus bypassing the difficulty of the optical pulse stretcher but allowing for maximum pulse energies to be delivered to the probe volume.

Higher pulse energies have additional benefits other than increased signal. With higher pulse energies, the overall signal gain on the intensifier can be reduced, therefore reducing a significant source of noise in the system. With high enough laser energies (or new CMOS camera technology), the high-speed intensifier could be removed entirely, though results are needed to understand the optimal signal collection strategy: low signal/low noise on a CMOS-only detection signal or high signal/higher noise on an intensified-CMOS detection system. One of the most critical needs in improving the high-speed Raman measurements are advances in CMOS-camera technology, namely in an increase in the dynamic range or bit-depth. The current generation of CMOS camera technology offers 12-bits of dynamic range. Turbulent flame conditions require measurements over a large range of fluctuating temperatures and species mole fractions. Ignoring variations in species mole fractions that occur within the flow field, there is a possibility of a ten-fold change in signal between the highest-temperature regions and cold air and fuel on every Raman detection channel due to number density and cross section effects. Since the calibration conditions were in cold flows of known composition, near the stoichiometric contour (high temperatures and low concentrations) where scalar data is critical, the measured signals used to calculate species mole fraction were often near the noise level of the camera. If new camera technology is not
immediately available, new signal processing and data processing methodologies need exploration to increase measurement signal-to-noise.
References


[123] Thorlabs, “Mounted Absorptive Neutral Density Filters Mounted Ø1 / 2 ‘
Absorptive Neutral Density Filters Mounted Absorptive Neutral Density Filters Add To Cart Mounted Ø25 mm Absorptive Neutral Density Filters Add To Cart

228


