DEVELOPMENT OF HIGH-SPEED LASER DIAGNOSTICS FOR THE STUDY OF ADVANCED PROPULSION SYSTEMS

BY

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DISSERTATION

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ABSTRACT

Combustion in next-generation high-speed propulsion systems involves highly turbulent reactive flow conditions, often beyond the limits of our physicochemical understanding. It is a challenge to produce reliable combustion as localized extinction and instabilities are highly transient in nature. It is in this effort that the progressing capabilities of laser diagnostics open new possibilities of exploring the chemistry and fluid dynamics of complex reacting flows. The primary objective of this study is to develop and apply kHz-rate planar imaging to further our understanding of ignition and flame dynamics in turbulent flames relevant to next-generation propulsion systems. High-repetition-rate planar laser-induced fluorescence (PLIF) is used to produce temporally resolved two-dimensional concentration fields of the molecular species targeted. Novel advancements in this study include kHz-rate hydroxyl radical (OH) PLIF of combustion phenomena within a supersonic wind tunnel cavity flameholder, the development and demonstration of a 50-kHz OH PLIF system using a small-scale turbulent flame, the development and demonstration of 10-kHz PLIF of the nitric oxide (NO) molecule, and the development and demonstration of a novel strategy for PLIF of the methylidyne (CH) radical at 10 kHz. The methods and diagnostics presented here are on the forefront of the field, extending the limits of the current capabilities to greater imaging rates and expanding the pool of molecular species that can be imaged at high rates.

The diagnostics methods are demonstrated in a range of laboratory combustors and propulsion systems. These include (1) an extensive study of a direct microwave plasma coupled flames developed for use in supersonic combustion systems, (2) high enthalpy supersonic combustors, and (3) high shear turbulent combustors. Laser diagnostics are used to examine direct-coupled, plasma ignited and sustained flames, for multiple flame types and nozzle geometries. OH radical number densities are quantified using PLIF and temperature measured by Rayleigh scattering thermometry. High-repetition-rate laser diagnostic methods are implemented to simultaneously record OH PLIF and chemiluminescence within the plasma-enhanced flame, allowing for
temporally resolved observation of OH radicals in the plane of the thin laser sheet as well as volume-integrated excited state emission. High-speed OH PLIF is also applied to a Mach 2 combustor to characterize the behavior of the flameholding cavity. The effects of cavity fueling rate are explored and discussed. High-shear turbulent combustors produce convoluted reaction zones, and approach the boundaries between theoretical combustion regimes. Visualization of CH radicals is an excellent way to probe the nature of such flows, and validate theory and computational models. Heretofore, CH PLIF framing rates have been constrained because the strategies have required very high laser pulse energies. In this work a new approach is presented, using the C-X (0,0) transition, which produces excellent signal even with the low laser pulse energies typical of kHz-rate Nd:YAG and dye laser systems. The novel capabilities of the diagnostics in this study will provide new insights that can help to optimize new combustor geometries and flame enhancement technologies in our future.
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I did not enter graduate school with a true appreciation of my great fortune. There was no examination of research groups and the professors that led them, or any list of schools and programs, ranked by my interest. In hindsight, this transition lacked much in the way of due diligence. And yet, I sincerely believe I landed in a rare and enviable position, one that, had I been more knowledgeable going into my graduate career, would have certainly be a top choice.

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CHAPTER 1 INTRODUCTION

1.1 Background and Motivation

Development of next generation high-speed propulsion systems is fraught with a vast range of technical challenges. The scramjet flowpath, for example, which offers the benefits of a simple design, reduced weight, low drag and high combustion efficiency, faces significant technical challenges in flow control and combustion stability due to the nature of supersonic airflow through the entire engine and the resulting high Reynolds number regimes [1, 2]. The impetus for this study is the strong need for a better understanding of ignition leading to stable combustion—and other combustion transient—in supersonic, air-breathing propulsion systems.

Laser-based methods have the ability to quantify the composition and thermodynamic state of an interrogation volume with high spatial and temporal resolution. Laser diagnostics and spectroscopy allow for in situ measurements of combustion processes without requiring the insertion of invasive probes. As such, these methods avoid disturbing the process under interrogation. Furthermore, laser spectroscopy can be utilized to study systems where probe-based techniques are impractical due to harsh conditions such as high temperatures, high velocities, corrosive reactants, etc. [3]. The data collected is invaluable in a thorough validation of combustion models. Traditional probe techniques for combustion measurements have been superseded by laser diagnostics that offer high spatial and temporal resolution, selective species targeting, sensitivity, and noninvasive nature [4].

Applying laser diagnostics to study high-enthalpy high-Reynolds reacting flows, like those that model a scramjet flowpath undoubtedly yields useful information, and many of these diagnostics have been extended to two dimensional measurements. However, there are short duration events such as ignition or blowoffs that are fast developing. Two-dimensional diagnostics that acquire
molecular concentration or temperature fields have generally been limited to acquisition rates of around 10 Hz. This may only yield a few frames of the event, with too much time between frames to correlate subsequent frames and so little is learned about the event unfolds. Recent advancements in laser and imaging technology have enabled high-speed diagnostics. Now, some of the same or comparable diagnostics can be performed at kHz to MHz rate. These methods and the equipment available continue to rapidly improve, providing additional information for propulsion system design and modeling.

In high Reynolds number turbulent flows, reliable flame ignition and stabilization is a major issue. One solution is to use electromagnetic energy from a non-equilibrium plasma. Plasma enhanced combustion explores the possibilities of improving thermal energy conversion through the selective deposition of electromagnetic energy to ignite and/or sustain oxidation chemistry [5, 6]. Electromagnetic energy and flame interaction have been utilized pursuant to a variety of goals, from a desire for increased combustion efficiency, faster reaction rates, and operation under harsh conditions. Demonstrated benefits include the reduction pollutants and fuel reformation, improved fuel efficiency, and more reliable and rapid ignition [7-10]. Other studies have observed increased flame stability and ignition at conditions beyond conventional combustion limits, including ultra-lean oxidation and high speed flow [11-13]. Plasma assisted combustion is a potential solution for engine operation in hypersonic flight, where complete and stable combustion is exceedingly difficult using traditional methods.

While the enhancement of a flame using electromagnetic energy has been well study and quantified, the exact mechanism(s) responsible for enhancement are still actively being explored. A number of studies have provided a glimpse into various aspects of this process [14-18]. Flame enhancement is likely a product of multiple physical and chemical plasma-flame interactions, including: (1) decomposition of the fuel from larger to smaller hydrocarbons and creation of
radicals via collision with electrons; (2) radiation-induced electron excitation; (3) increased flame temperature by ohmic heating; (4) production of excited state species, ions and electrons; and (5) in-situ fuel reformation. To date, a large number of different discharge systems including microwave, DC, RF, etc. have been investigated by various research groups [19-25] using both experimental and numerical approaches. Nonetheless, the prospect of gaining insight into the synergistic integration of electrodynamics and combustion chemistry is a formidable challenge and requires further work. An extensive review can be found in ref. [5].
1.2 Scope of Research

1.2.1 Laser Diagnostics

The aim of this study is to develop and advance kHz-rate combustion diagnostics with the purpose of improving our capability to understand turbulent combustion physics, make meaningful measurements of high-enthalpy flows, and design robust and reliable super- and hyper-sonic engines. Imaging of combustion radicals has been a cornerstone diagnostic of the field for the past two decades, allowing researchers to visualize flame structure and behavior, but has been limited to low-frequency snapshots. Increasing the frame rate and expanding the pool of detectable species for high-speed planar-laser induced fluorescence (PLIF) is the primary focus of this study, which includes significant advancements in the state of the art in combustion diagnostics for the study of turbulent combustion. The following paragraphs describe distinct investigations.

The results of an experimental investigation of a supersonic cavity combustor using high-frequency planar imaging diagnostics are presented in this work. Specifically, 10-kHz OH (PLIF) is used to capture flame geometry and dynamics within a cavity flameholder in a supersonic crossflow (Mach 2). The combustor behavior is examined for direct fuel injection into the cavity over a range of fuel flow rates. The 10-kHz PLIF used here is the first integration of sustained high-frequency PLIF imaging in a supersonic combustor, and while the frame rate is not sufficiently fast to resolve flame dynamics in general, these measurements allow the acquisition of a large and statistically significant database and represent a first step towards time resolution within a high-speed flowfield.
This work includes a demonstration of the first use of OH PLIF at a rate of 50 kHz using a continuously operating laser system, where the only limitation for data record length is the camera memory size. An Nd:YAG-pumped dye laser system is used for OH excitation and an intensified CMOS camera for fluorescence imaging. Results are presented for a stable Hencken burner flame and a turbulent flame produced by a transient-arc DC plasmatron.

The high-repetition rate PLIF imaging of both cold (~300 K) and hot (~2400 K) nitric oxide (NO) at a framing rate of 10 kHz is investigated. The laser system is composed of a frequency-doubled dye laser pumped by the third harmonic of a 10-kHz Nd:YAG laser to generate continuously pulsed laser radiation at 226 nm for excitation of NO. The LIF signal is detected using a high framerate, intensified CMOS camera, yielding a continuous cinematographic propagation of the NO plume where data acquisition duration is limited only by camera memory. The pulse energy of the beam is ~20 μJ with a spectral width ~0.15 cm⁻¹, though energies as high as 40 μJ were generated. The estimated signal-to-noise ratio (SNR) for the cold seeded flow and air plasma exceed 50 with expected NO concentrations of 6000 to 8000 parts per million (ppm, volume basis). Images show distinct, high-contrast boundaries. The plasma-assisted flame images have a SNR of less than 10 for concentrations near 1000 ppm. For many combustion applications, the pulse energy is insufficient for PLIF measurements. However, the equipment and strategies herein could be applied to high-frequency line-imaging of NO at concentrations of 10 to 100 ppm. Generation of 226-nm radiation was also performed using sum-frequency mixing (SFM) of the 532-nm-pumped dye laser and 355-nm Nd:YAG third harmonic but was limited in energy to 14 μJ. Frequency tripling a 532-nm-pumped dye laser produced 226-nm radiation at energies comparable to the 355-nm pumping scheme.

A novel strategy for PLIF of the methylidyne (CH) radical at kHz-rates was explored. It is demonstrated that the C-X(0,0) band is a good option for high-speed CH PLIF due to its strength
and the convenience of the excitation wavelength ($\lambda = 314$ nm) that can be generated by a dye laser operating with red dye. Within this band there is significant interference from hydroxyl (OH) fluorescence. However, this can be avoided by careful wavelength selection. The strength of the transition is very favorable for continuous kHz-rate laser systems which produce low pulse energies ($E_p < 1$ mJ). The method is first demonstrated using a 10-Hz system to demonstrate its potential. The study then moves to 10-kHz operation in laminar and turbulent CH$_4$-air flames.

1.2.2 Plasma Enhanced Combustion

As part of the effort to improve combustion performance in high-Reynolds number flows, investigations of plasma enhanced combustion have been performed.

A tunable microwave waveguide is used to initiate and enhance combustion by coupling an atmospheric plasma discharge to a premixed methane/air flame. The absorbed microwave power ranges from 60 to 150 W, generated from a continuous source operating at 2.45 GHz, whereas combustion power ranges from 200 to 1000 W. OH radical number densities were measured using planar laser-induced fluorescence (PLIF) and temperatures were measured using Rayleigh scattering thermometry for various flow rates, equivalence ratios, and power levels. Increases in reaction volume, OH density, and temperature were observed as power increased. Premixed and non-premixed flames are studied, using both solid and hollow inner conductors in the plasma applicator nozzle. Plasma power is controlled by adjusting the reflected microwave power, measured at a dummy load attached to a circulator. Maximum OH radical number densities were quantified as approximately $(3-5) \times 10^{16}$ cm$^{-3}$ for plasma powers around 100 W, with small variation between configurations. The maximum temperatures occurred in the non-premixed flame, where the plasma is generated in air, reaching values of 3500 K. Temperatures are lower, peaking at 2000 K, when the plasma is generated at the air-fuel boundary or the air-premixed
boundary through use of the hollow inner conductor. Additional parameters are adjusted, including flow rates, power level, equivalence ratio, and the effects are discussed. Non-premixed configurations are ill-suited for flame enhancement, whereas a premixed flow through the hollow electrode best demonstrates non-thermal plasma assisted combustion.

A second study utilizes a frequency-doubled, diode-pumped, 10-kHz Nd:YAG laser to pump a frequency-doubled, high-speed tunable dye laser to excite OH radicals in a plasma-enhanced flame. Fluorescence and chemiluminescence from a microwave-plasma-ignited and stabilized flame are simultaneously recorded by intensified CMOS detectors at 10,000 frames per second (fps) to yield new understanding of the plasma-assisted flameholding mechanism and allow tracking of individual flow features.
1.3 Dissertation Overview

There are 8 chapters in this dissertation, with chapters 4 to 7 documenting four distinct areas of interest, and a total of seven independent experiments distributed through those four chapters.

Chapter 2 contains a brief explanation of the theory behind the laser diagnostics used in the study. The primary focus is PLIF theory, as this diagnostic dominates the total body of work.

Chapter 3 describes the equipment used in these experiments. This includes a brief background on laser principles and the Nd:YAG and dye laser systems used throughout the work. Cameras and intensifiers are discussed, as well as simpler photodetectors. Finally, the plasma torches and supersonic wind tunnel are introduced.

Chapter 4 starts the experimental portion of the paper with a investigation of plasma assisted combustion using a microwave torch using 10-Hz diagnostics.

Chapter 5 contains three experiments utilizing high-speed OH PLIF. Section 5.1 reports 10-kHz measurements of a cavity flame holder in a supersonic wind tunnel. In Section 5.2, the first application of continuous 50-kHz OH PLIF is demonstrated. Section 5.3 documents measurements and findings of simultaneous OH PLIF and OH* chemiluminescence of the microwave torch from Chapter 4 at a repetition rate of 10 kHz.

Chapter 6 is limited to one study involving 10 kHz-rate NO PLIF in hot and cold flows

Chapter 7 contains an exploration of a novel pumping strategy for CH PLIF that is advantageous for high-speed diagnostics. This is split into two sections: in section 7.1 the method is proofed and investigated using a conventional 10 Hz system, and in section 7.2 the method is applied with a 10-kHz system.
CHAPTER 2  LASER DIAGNOSTICS FOR COMBUSTION

2.1 Laser-Induced Fluorescence

Laser-induced fluorescence (LIF) is a common and well-established method for the detection of molecules or interrogation of thermodynamic state. LIF is highly selective by quantum state, and quantum mechanics governs the absorption and emission of radiation. The method is often used because it can be species selective, but it also is affected by collisional energy transfer and so can indicate thermodynamic properties such as temperature, pressure, and velocity. The combustion community has made great use of the ability to detect minor species in flames at concentrations below 1 ppm [4, 26-28]. LIF can be extended to a two-dimensional measurement known as planar laser-induced fluorescence (PLIF), wherein a concentration field of the target species is imaged [29]. This allows for greater determination of the spatial structure of a reacting flow [30]. The target species may be chosen because its occurrence marks the reaction zone or combustion products, as is often the case for OH and CH PLIF [31]. The target species may also be a molecule like NO, seeded into the flow for visualization of fluid motion [32].

2.1.1 Two-level model

Laser-induced fluorescence is fundamentally the absorption of applied laser radiation, producing excited state molecules or atoms that quickly return to thermal equilibrium by spontaneous emission. The changes in energy, or transitions, are limited to molecules in a quantum state suitable for absorption of radiation. The emission of radiation is also limited by selection rules to a distribution of transitions. LIF can be first introduced as a two-level system with an upper and lower electronic state.
Figure 2.1 illustrates the two-level system and the mechanisms for energy transfer. This is a greatly simplified representation of the actual physics, but is a useful model for developing a mathematical model of a steady-state system. There are four mechanisms for energy transfer in the model. Absorption of radiation promotes the molecule or atom from the ground electronic state \( E_1 \) to an excited electronic state \( E_2 \) with a rate constant of \( b_{12} \). The molecule may be perturbed by a radiation field and emit a resonant photon, returning to the ground state with a rate constant of \( b_{21} \). Excited state molecules can also relax through collisions with other molecules, a radiationless process called quenching characterized by the rate constant \( Q_{21} \). The reverse process, collisional excitation, occurs at a negligible rate relative to other transfer mechanisms and so is typically ignored and the collisional quenching rate constant simplified to \( Q \). The last mechanism in this simple model is spontaneous emission, wherein a high energy molecule spontaneously emits radiation as it relaxes to the lower electronic energy level. This mechanism is also called fluorescence and occurs at a rate of \( A_{21} \). Further details of these types of energy transfer follow. The rate constants \( b, A, Q, P, \) and \( W \) are occasionally incorrectly referred to as rates in other works. The nomenclature used by Eckbreth [33] is followed in this work, where we adhere to the standard nomenclature in chemistry and refer to these terms as rate constants [34].
Figure 2.1 A diagram of a two-level LIF model with energy transfer processes marked by their rate constant. Curvy arrows are used for transfer processes that involve electromagnetic radiation exchange while straight lines denote radiationless processes.

Absorption (b_{12}) is the event where energy from radiation is captured and added to a molecule’s internal energy, resulting in an excited state. This excitation process removes molecules from the ground state population and increases the upper state population. The rate is directly proportional to the Einstein transition probability B_{12}, and can be described by the equation

$$b_{12} = \frac{B_{12} I_\nu}{c}$$  \hspace{1cm} (2.1)

where I_\nu is the incident laser irradiance per unit frequency, with units W/cm^2 \cdot sec^{-1}, and c is the speed of light. A more rigorous model requires accounting for laser lineshape effect [35-39], as described by
\[ b_{12} = \frac{B_{12}}{c} \int \nu (\nu) g(\nu) d\nu \quad (2.2) \]

where \( I_{\nu}(\nu) \) is the laser lineshape and \( g(\nu) \) the absorption lineshape. The incident laser irradiance can be decomposed such that

\[ b_{12} = \frac{B_{12}}{c} I_{\nu}^{0} \int \nu (\nu) g(\nu) d\nu \quad (2.3) \]

where \( I_{\nu}^{0} \) is the normalized spectral irradiance and \( L_{\nu}(\nu) \) is the spectral distribution function. The integral of the two distributions is the line overlap integral and is a dimensionless number \( \leq 1 \). If the laser lineshape is sufficiently broad such that it varies negligibly over the absorption linewidth, then the overlap integral is 1 and Eq. 2.1 is valid so long as \( \nu \) is the laser line center.

**Stimulated Emission** \((b_{12})\) is the release of radiation induced by the interaction of the molecule with incident radiation. The rate is directly proportional to the Einstein transition probability \( B_{21} \), and can be described by the equation

\[ b_{21} = \frac{B_{21}}{c} I_{\nu} \quad (2.4) \]

where \( I_{\nu} \) is the incident laser irradiance per unit frequency, with units \( \text{W/cm}^2 \cdot \text{sec}^{-1} \), and \( c \) is the speed of light. Stimulated emission is the reverse of absorption, and there is a fixed relationship of the transition probabilities, described as

\[ g_{1}B_{21} = g_{2}B_{12} \quad (2.5) \]

where \( g_{1} \) and \( g_{2} \) are the degeneracies of state 1 and 2, respectively [40].
**Spontaneous Emission** \( (A_{21}) \) is the unprompted release of radiation associated with the de-excitation of the molecule from state 2 to state 1. This radiation is the fluorescence signal sought in LIF experiments. The Einstein coefficient for spontaneous emission is the rate constant \( A_{21} \), and is related to the stimulated emission rate constant

\[
\frac{A_{21}}{B_{21}} = \frac{8\pi h \nu^3}{c^2}
\]  

where \( h \) is Planck’s constant and \( \nu \) is the wavenumber corresponding to the \( 2 \rightarrow 1 \) transition. The relationship between \( A_{21} \) and the absorption rate constant is clearly

\[
\frac{A_{21}}{B_{12}} = \frac{g_1}{g_2} \frac{8\pi h \nu^3}{c^2}
\]  

**Collisional quenching or electronic quenching** \( (Q_{21}) \) occurs when the excited molecule collides with another molecule and loses enough energy in the exchange to move to state 1. For more complex models, this definition will require refinement to a change in the electronic state due to collisions, and collisions only affecting rotational and vibrational states will be dealt with separately. There is no emission of radiation when a molecule leaves state 2 through collisional quenching. The quenching rate constant is a function of the collisional partner species and its energy, and so \( Q_{21} \) is dependent on gas composition and temperature. Quantitative measurements require the evaluation of \( Q_{21} \), and this dependency causes great difficulty combustion features rapid and drastic changes in composition and temperature. This can be expressed as

\[
Q = \frac{P}{kT} \sum_j x_j \nu_j \sigma_{Q,j}
\]
where \( x_j \) is the mole fraction of collisional partner species \( j \), and \( v_j \) is the relative velocity of the two colliding molecules, and \( \sigma_{Q,j} \) is the quenching cross section for species \( j \). The quenching rate constant is proportional to \( p \), pressure, and inversely proportional to \( T \), temperature. In high pressure combustors, quenching rates become the dominant mechanism for de-excitation.

**Predissociation (P)** is when the energy absorbed by a molecule begins to decompose the molecule into separate species. The predissociation rate constant \( P \) is an intrinsic molecular property and specific to an excitation transition.

**Photoionization (\( W_{2i} \))** occurs when a molecule absorbs sufficient radiation to excite an electronic to an unbound state, leaving the molecule ionized. The photoionization rate constant \( W_{2i} \) from energy level 2 can be written

\[
W_{21} = \frac{\sigma_{2i} I_i}{h \nu_i}
\]

(2.9)

where \( \sigma_{2i} \) is the photoionization cross section from energy level 2, \( I_i \) is the irradiance of the photoionizing laser, and \( \nu_i \) is the frequency.

All the rate constants are per molecule, and the rate can be found by multiplying the rate constant by the population of the starting state. The rate of population change for state 1 and 2 for this two level system can be written

\[
\dot{N}_1 = -N_1 (b_{12}) + N_1 (b_{21} + A_{21} + Q_{21})
\]

(2.10)

\[
\dot{N}_2 = N_1 (b_{12}) - N_2 (b_{21} + A_{21} + Q_{21} + P + W_{2i})
\]

(2.11)

where \( N_i \) is the population of state \( i \) and \( \dot{N}_i \) is the rate of change of population \( i \). After reaching an excited state through absorption (\( b_{12} \)), molecules can leave \( N_2 \) through spontaneous emission.
(A_{21}), or by the competing mechanisms of stimulated emission (b_{21}), collisional quenching (Q_{21}), predissociation (P), and photoionization (W_{2i}).

Typically, significant photoionization only occurs by design, and most excited states are not predissociative. If predissociation and photoionization are negligible, then there is only population exchange between state 1 and 2. This results in a closed system where

\[ \dot{N}_1 + \dot{N}_2 = 0 \]

and if the initial excited state population is negligible,

\[ N_1 + N_2 = \text{constant} = N_1^0 \]

with \( N_1^0 \) referring to population of state 1 prior to laser excitation. By solving Eq.2.11 the population \( N_2 \) as a function of time can be found

\[ N_2(t) = \frac{b_{12} N_1^0}{r} \left(1 - e^{-rt}\right) \]

where \( r = b_{12} + b_{21} + A_{21} + Q_{21} \). By defining a saturation irradiance

\[ I_{\text{sat}} = \frac{A_{21} + Q_{21}}{B_{12} + B_{21}} \cdot c \]

and rearranging, Eq. 2.14 can be equivalently written

\[ N_2 = N_1^0 \frac{B_{12}}{B_{12} + B_{21}} \frac{A_{12}}{1 + \frac{I_{\text{sat}}}{I_{\nu}}} \]

The fluorescence power \( F \) is the product of the population and spontaneous emission rate constant of the excited state (\( N_2 A_{21} \)); the photon energy of the frequency of the fluorescence
(hv); and the geometrical parameters: collection solid angle (Ω), laser beam focal area (A), and the pathlength of the observed fluorescence (l). Substituting for \( N_2 \) using Eq.2.16, the fluorescent signal power can be expressed

\[
F = h\nu N_2 A_{12} \frac{\Omega}{4\pi} l A = h\nu A_0 \frac{\Omega}{4\pi} l A N_1^0 \frac{B_{12}}{B_{12} + B_{21}} \frac{A_{21}}{I_{sat}} \frac{I_{sat}}{I_v}
\]

(2.17)

The unknown variable \( N_1^0 \) is a fraction of the total species population and determined through the Boltzmann fraction for \( N_1 \) at the species temperature.

**2.1.2 Vibrational and Rotational States**

The energy states of diatomic molecules can be described by a hierarchy of energetic components. The first component is the electronic energy level, the second is vibrational, and the third is rotational. Internal energy of the molecule is the sum of these three components, and transitions involving a change in the electronic state are called rovibronic, indicating the change in all three components. Excitation of the molecule by a narrowband laser is targeted to a single rovibronic transition. Figure 2.2 illustrates excitation of a molecule and the hierarchy of energy components.

While the excitation transition is between two defined rovibronic states, the subsequent spontaneous emission occurs over many transitions. The excited state molecule may fall to a range of rotational and vibrational energy levels. Additionally, collisions between molecules can occur before emission, changing the rotational, vibrational, or even electronic energy level to another energetic state. Rotational energy transfer (RET) and vibrational energy transfer (VET) become increasingly important at high pressures, due to the increase in collisional rates. Figure
2.3 illustrates spontaneous emission of an excited molecule, producing fluorescence at a range of wavelengths.

**Figure 2.2** Excitation diagram. The molecule absorbs a photon equal to the energy gap between the upper and lower states. Within each electronic potential well are multiple vibrational levels and each vibrational level contains multiple rotational levels.
Figure 2.3 Fluorescence diagram. The molecule emits a photon equal to the energy gap between the upper and lower states but does not have to fall through the excitation transition. Excited molecules may fall to the ground vibrational state or the first vibrational level. An analogous process occurs with respect to the rotational transition. Furthermore, both the vibrational and rotational states may change before fluorescence occurs due to collisions. This is called vibrational energy transfer (VET) and rotational energy transfer (RET).

2.1.3 Linear Regime

When the excitation laser energy is well below the saturation limit, Eq. 2.17 can be simplified to

\[ F = \frac{h \nu}{c} I A N_1^0 B_{12} I_v \frac{A_{21}}{A_{21} + \tilde{Q}}. \]  

\[ (2.18) \]
Under this condition \((I_v << I_{\text{sat}})\), the fluorescence signal power \(F\) is proportional to the incident laser irradiance \(I_v\) and this is termed the linear regime. The term \(\frac{A_{21}}{(A_{21} + Q)}\) is the fluorescence efficiency and is typically much less than one. Quenching is the dominant relaxation path for molecules that have absorbed laser energy. To make quantitative measurements in the linear regime, the quenching rate needs to be accounted for. This is a challenging task; the quenching rate constant is a function of the energy exchange between colliding molecules. It is a function of both gas mixture composition and temperature, two parameters that are not trivial to measure and naturally have steep gradients in a reacting flow. For a laminar flame with well understood chemistry, quenching corrections can be determined adequately. Other avenues that sidestep quenching rate corrections are preferred for unsteady and turbulent flames.

\[
F = \frac{h \nu c}{I} A N_i^0 \frac{B_{12}}{B_{12} + B_{21}} A_{21} \tag{2.19}
\]
Notice that the fluorescence power $F$ is now independent of laser irradiance $I$. The combination of being independent from laser power, avoiding the difficulties of quenching rate corrections, and maximizing the fluorescence signal, all are attractive attributes of saturated LIF. However, this fully saturated condition would require a laser pulse with infinitely short rise and fall times, and infinitely steep spatial gradients. The pulse would have to be a perfect step function of energy, both spatially and temporally, and because this is far from reality, there is always at least some unsaturated (and therefore linear) LIF at the leading and trailing edges, and on the sides of the beam. There is also the issue of energy exchange within the rotational manifold. More comprehensive models and methods than presented here do try to address these challenges.

In laser-induced predissociative fluorescence (LIPF), an excitation transition to a highly predissociative state is selected such that the predissociative rate constant is much greater than the quenching rate constant ($P >> Q$) [41, 42]. Photoionization controlled-loss spectroscopy (PICLS) is similar to LIPF, but it is the photoionization rate that dominates the loss mechanisms [43, 44]. Both of these methods introduce a loss mechanism large enough to make quenching insignificant while being easier to calculate and account for. The disadvantage is the decrease in fluorescence power this causes, reducing species detectivity.

### 2.1.5 Applications of High-Speed Laser Diagnostics

Early high-repetition-rate laser diagnostics for combustion used excimer lasers [45] or double-pulsed Nd:YAG lasers [46, 47]. The first demonstration of planar laser-induced fluorescence (PLIF) at kHz rates over more than a pair of images utilized four individual, double-pulsed Nd:YAG lasers pumping a commercial dye laser (to produce a burst of eight pulses) and an array of eight independent intensified charge-couple device (CCD) detectors to record the fluorescence. The OH field was thus imaged at a rate of 8 kHz and then 33 kHz in combination
with additional planar diagnostics [48, 49]. Other approaches for OH PLIF, however, are feasible, and more recently the use of a tunable, frequency-tripled Yb:YAG disk laser and intensified complementary metal-oxide-semiconductor (CMOS) detector have been demonstrated [50]. Combining the CMOS camera with diode-pumped solid-state lasers has spurred continued improvements in repetition rate and provided an opportunity for long-duration measurements not limited to a few images [51]. A pulse-burst laser (PBL) produces an envelope of high-repetition-rate pulses from i) a CW laser sliced by a Pockels Cell [52], ii) a Q-switched oscillator [53, 54], iii) a cluster of multiple lasers [55], or iv) a doped fiber laser [56, 57]. The pulse train can be amplified in several stages and wavelength tunability can be added with an optical parametric oscillator (OPO) [58]. Such systems are well suited for scattering-based measurements, where high pulse energies negate the need for an image intensifier [59]. OH PLIF has also been demonstrated using a PBL-pumped, injection-seeded, OPO at a rate of 50 kHz that produced over 100 pulses and images [60].

The recent development of high-speed tunable dye lasers and intensifiers creates the opportunity for selective tuning and sustained operation like that of the conventional 10 Hz systems. Such a system, complemented by additional lasers and cameras for simultaneous stereoscopic PIV, provides the tools for the study of transient phenomena in turbulent combustion through time-resolved flamefront tracking and three-component flowfield construction. These techniques have been demonstrated for jet diffusion flames [61-66], opposed jet flames [51, 67], and swirl-stabilized gas turbine combustion [63, 64, 68-70] at framing rates of 1.5 to 10 kHz, capturing the great significance of quasi-four dimensional flowfield measurements on flame stabilization investigation.
CHAPTER 3  EQUIPMENT AND FACILITIES

3.1 Laser Systems

Lasers provide a radiation source with well-defined attributes and adjustable parameters. The term laser is an acronym for Light Amplification of Stimulated Emission of Radiation. The word “light” refers to region of the electromagnetic spectrum from infrared through ultraviolet which includes visible radiation. Lasers contain an oscillator, which allows for the generation of photons by stimulated emission that are in phase, monochromatic, and directional, and that can be easily amplified.

Radiation can be absorbed or emitted through changes in the energy level of atoms. These energy levels are discrete, and the radiation absorbed or emitted must match the energy difference between the initial and final level. This bundle of electromagnetic radiation is called a photon. The transition of an atom from one energy level to another by interaction with photons can be described three ways: absorption, spontaneous emission, and stimulated emission. Absorption is the transition from a lower energy level to a higher level as the result of the interaction with a photon with the energy matching the energy difference of the two levels. Once absorbed, the electromagnetic energy no longer travels as radiation, but has been transferred into an atom with increased energy. Spontaneous emission is the reverse process. An atom radiates energy away as a photon, and drops to a lower energy level. An atom at a high energy level can also be stimulated by a photon to emit another photon of the same energy and direction. Stimulated emission also results in photons being in phase, meaning that the oscillation of the electromagnetic dipoles is synchronized between the photons.
The lowest energy level of an atom is referred to as the ground state, $E_1$. In general, almost all atoms are in the ground state, and each higher energy level has contains a lower number of atoms as the previous level. The rate at which atoms will transition to a higher energy level $E_2$ is dependent on the transition probability and by the difference in population of the initial and final energy levels. The rate of a transition from $E_1$ to $E_2$, will be greater that the rate of a transition from $E_2$ to $E_1$. This is because the atom is the population of the ground state, $N_1$, is much greater than the population of the higher energy excited state, $N_2$. This means an atom will have a higher rate of absorption than of emission. However, if a collection of atoms is supplied with a great deal of energy, it is possible to excite enough atoms so that the $N_2$ is equal to $N_1$, and the rate of absorption and rate of emission will be equal. $N_2$ cannot be greater than $N_1$ because the atoms are gaining and releasing photons at equal rates. Moreover, a further increase in $N_2$ would result in a rate of emission higher than the rate of absorption.

If three or more different energy levels are consider, it is possible to produce a population of a higher energy level greater than the population of a lower energy level. This is done by pumping atoms, meaning supplying radiation to increase the atoms’ energy level, from a ground state $E_1$ to an excited state $E_3$. Atoms at $E_3$ can release energy to arrive at $E_2$. $E_3$ is often an unstable state, such that the atoms drop $E_3$ to $E_2$ quickly. If supplied with enough energy, the system can reach a point where the population $N_2$ is equal to $N_1$, and $N_2$ can even exceed $N_1$. The limitation of the two-level system has been removed by pumping the atoms to the unstable $E_3$ instead of $E_2$. This is called a population inversion. A population inversion can be created more easily by using a four level system. This involves two unstable states, $E_4$ and $E_2$. When an atom is pumped from $E_1$ to $E_4$, it quickly decays to $E_3$ where it is more stable. Over time, atoms at $E_3$ will emit a photon, dropping to energy level $E_2$. $E_2$ is also unstable, so the atom quickly returns to $E_1$. The
result is an immediate population inversion between $E_3$ and $E_2$. Nd:YAG lasers operate in such a scheme.

### 3.1.1 Nd:YAG Lasers

The Neodymium-doped Yttrium Aluminum Garnet (Nd:YAG) laser is a common tool in laser diagnostics. It is a solid-state laser, with the lasing medium comprised of a YAG glass rod or slab with about one percent of the yttrium atoms replaced with Nd$^{3+}$ (triple ionized, or trivalent, neodymium) atoms. Often, these lasers are pulsed and contain a Q-switch to generate short pulse lengths. Nd:YAG lasers high intensity broadband radiation, from flash lamps or diodes, to excite the neodymium in the YAG rods, producing a population inversion. The Q-switch prevents gain of any spontaneous emission from the rods while this inversion builds, then suddenly permits feedback and gain of light through stimulated emission. The radiation from an Nd:YAG laser is infrared, 1064 nm. Nonlinear optics are often used to generate harmonics of the fundamental beam, producing radiation at 532 nm, 355 nm, and 266 nm.

An Nd:YAG laser uses flashlamps or diodes as a radiation source to pump neodymium atoms from the ground state to an unstable group of high energy levels. The atoms quickly drop to a more stable state denoted $^4F_{3/2}$, where they remain for about a quarter of a second. From there, the most likely transition is to the $^4I_{11/2}$ state, and the photons released from this transition have a wavelength of 1064 nm. The $^4I_{11/2}$ state quickly decays to the ground state $^4I_{9/2}$ so that a population inversion occurs between the upper state, $^4F_{3/2}$, and the lower state, $^4I_{11/2}$ [71].

Figure 3.1 illustrates the energy level scheme for an Nd:YAG laser. There are competing transitions for decay from the $^4F_{3/2}$ state, producing wavelengths other than 1064 nm, but they have a higher threshold and a lower gain.
With the laser medium in a state of population inversion, the rate of emission is greater than the rate of absorption, establishing an active medium. Light that passes through the active medium is amplified by inducing stimulated emission. The laser is formed by placing mirrors on either end of the medium, creating a resonant cavity. Lasing begins with spontaneous emission from an active medium. The emitted photon stimulates emission from excited state atoms, resulting in the release of an additional photon of the same phase, direction and energy (and therefore wavelength). This continues until equilibrium between excitation and emission is reached. All but a small fraction of the photons are contained within the cavity, traveling back and forth as

\[ \text{Figure 3.1} \quad \text{A four-level model for lasing. A population inversion is achieved between E3 and E2 when an external source pumps the ground state atoms to a unstable excited state E4, which quickly decays to E3 while any atom in E2 quickly decay to E1. Also shown are the equivalent energy levels for the Nd}^3+ \text{ in an Nd:YAG laser.} \]
they are reflected off the cavity end mirrors, producing a standing wave. The laser beam is formed by having one of the mirrors permits a small fraction of the light to escape. This partially reflective mirror is called the output coupler. The laser pulse will continue as long as the excitation source (e.g. flashlamp) is active and producing the population inversion.

It is often more useful to have the laser pulse energy contained in a shorter pulse length, such that the peak power is greater. The short pulse length is useful for improving temporal resolution of fast events, and the high peak power is better suited for nonlinear phenomena, such as frequency mixing and dye laser pumping. This can be achieved by using a Q-switch within laser cavity, between one end of the laser medium and the highly reflective mirror, as diagrammed in Figure 3.2. A Q-switch allows for fast control of the cavity gain, making it possible to prevent lasing while the excitation source continues to build up a population inversion.

Three components create a Q-switch: a polarizer, a Pockels cell, and a quarter-wave plate. Initially, the Q-switch is in a closed state, and vertically polarized light exiting the medium passes through the polarizer. The Pockels cell contains a nonlinear crystal, typically potassium dideuterium phosphate (KD*P) in Nd:YAG cavities, that acts as a variable wave plate when a high voltage potential is applied. In the closed state, no voltage is applied to the Pockels cell and there is no change in the polarization of the light passing through. The vertically polarized light then encounters the quarter-wave plate and becomes circularly polarized. After reflecting off the end mirror, the light again passes through the quarter wave plate, resulting in horizontal polarization, and through the Pockels cell. The horizontally polarized light is no longer passes through the polarizer, and is now reflected out of the cavity. The closed Q-switch produces high cavity loss by preventing oscillations in the cavity. In this state, the population inversion can continue to grow in strength while the excitation source continues to irradiate the medium.
Figure 3.2 Diagram of a basic Q-switch. The Q-switch is located between the lasing medium and the cavity end mirror and is comprised of three elements: a quarter-wave plate, a Pockels cell, and a polarizer. A voltage potential across the Pockels cell controls cavity feedback.

There is a point where the population inversion is maximized, about 200 us for a flashlamp pump Nd:YAG, and the Q-switch is opened by applying fast high-voltage pulse to the Pockels cell. By applying the proper potential, the Pockels cell now acts as a quarter-wave plate. Vertically polarized light exiting the medium is now circularly polarized by the Pockels cell, and then horizontally polarized by the quarter-wave plate. After reflecting off of the end mirror, the light is again circularly polarized by the quarter-wave plate then vertically polarized by the Pockels cell. Thus, with voltage applied, the light is returned to its original vertical polarization after passing through the Pockels cell and quarter-wave and back. The vertically polarized light is permitted through the polarizer and back into the active medium, where it stimulates emission of more photons. The open Q-switch returns the cavity to a low loss state.

The Nd:YAG lasers used in this work can be divided into two groups based on their pulse repetition rate. For low-speed diagnostics, conventional flashlamp pumped laser operating at 10 Hz are used. For high-speed diagnostics, recently developed diode pumped lasers operating at 10 kHz to 50 kHz are used. Both types of lasers operate using the fundamental physics and
mechanisms described in the previous sections. Table 3.1 contains all the Nd:YAG lasers used, and notes which experiments each was part of.

Table 3.1 The Nd:YAG and dye lasers used in this work.

<table>
<thead>
<tr>
<th>Series</th>
<th>Quanta-Ray</th>
<th>INNOSLAB</th>
</tr>
</thead>
<tbody>
<tr>
<td>Manufacturer</td>
<td>Spectra-Physics</td>
<td>EdgeWave GmbH</td>
</tr>
<tr>
<td>Lasing medium</td>
<td>Nd:YAG rod</td>
<td>Nd:YAG slab</td>
</tr>
<tr>
<td>Pump source</td>
<td>Flash lamps</td>
<td>Diode laser stacks</td>
</tr>
<tr>
<td>Dye laser series</td>
<td>Lumonics Hyperdye HD-300</td>
<td>Sirah Lasertechnik CREDO</td>
</tr>
<tr>
<td>Repetition rate</td>
<td>10 (2-15) Hz</td>
<td>10 (4-12) kHz</td>
</tr>
<tr>
<td>Model</td>
<td>GCR-150</td>
<td>GCR-170</td>
</tr>
<tr>
<td>Max power (max, 532 nm)</td>
<td>3.0 W</td>
<td>4.5 W</td>
</tr>
<tr>
<td>Pulse energy (max, 532 nm)</td>
<td>300 mJ</td>
<td>450 mJ</td>
</tr>
<tr>
<td>Section</td>
<td>4.1</td>
<td>7.1</td>
</tr>
</tbody>
</table>

3.1.2 Dye Lasers

The utility of dye lasers stems from the broadband fluorescence of the dye medium and the oscillator cavity grating. In combination, these two factors create a narrowband, wavelength-tunable laser. The lasing medium consists of organic dye dissolved in a liquid solvent, with the solution typically circulated through a quartz dye cell. The dye molecules are large, containing numerous bonds and degrees of freedom of motion. This creates a vast number or finely spaced
energy levels over a broad range, including many vibrational sublevels within the electronic states, allowing the dye to provide gain over a large span of wavelengths. Interaction between dye molecules and the organic solvent cause collisional broadening and a smearing of the discrete transition energies into a continuous absorption curve with a width of 50-100 nm. Different families and subtypes of dye are available so that dye lasers can cover a large range of usable wavelengths with the appropriate dye selection, from the infrared to the ultraviolet. The more efficient visible range dyes are often used to generate ultraviolet output using frequency doubling and mixing.

When the dye absorbs a photon, the dye molecules are pump from the ground singlet state $S_0$ to the higher vibrational levels of the first excited singlet state $S_1$. Collisions between dye molecules and the solvent molecules cause the dye molecules to drop in energy to the first vibrational $v_0$ level of the $S_1$ state. This transfer of energy is radiationless, and causes the population of the higher vibrational levels to remain low relative to the molecules at the ground state $S_0$. The emission of radiation from the dye occurs as the dye molecules at $v_0$ of $S_1$ relax to the practically unpopulated higher vibrational levels $v_h$ of $S_0$. The population of $v_h$ of $S_0$ remains near zero because the molecules quickly drop to $v_0$ of $S_0$ through collisions with the solvent molecules. With enough pumping energy, a population inversion is produced between the lowest vibrational level $v_0$ of the $S_1$ state, and the higher vibrational levels $v_h$ of the ground singlet state $S_0$. As with the Nd:YAG lasers, dye laser pumping scheme can be described by a four-level model.

Reflectors are placed at either end of the lasing medium to form a cavity. To produce a narrowband dye laser, a dispersive element, often a grating, is used at one end of the cavity. The light reflects off or refracts through the dispersive element at an angle that is a function of wavelength. Only a small range of angles will send the light back into the cavity, and so only
photons of a narrow range of wavelengths can oscillate in the cavity. Rotation of the dispersive element changes the wavelength selection, making the laser tunable.

In these experiments, the dye is pumped by the output of a pulsed Nd:YAG laser after either frequency doubling to produce 532-nm radiation, or frequency tripling to produce 355-nm radiation. The Nd:YAG beam passes through optics to stretch the beam horizontally before it is focused in the vertical direction onto the dye cell, resulting in a thin line of pumped dye solution. This is the active medium of the dye laser. The resonant cavity is formed with an output coupler beside the dye cell, and a grating at the other end. Between the dye cell and the grating there is a beam expander and a mirror. The grating is mounted to a rotary drive for wavelength tuning. Only a portion of the pumping beam is directed onto the resonator dye cell. The majority is focused onto one or more amplifier cells, producing an active medium in the path of the laser beam. Table 3.2 describes the laser dyes used in this study.

<table>
<thead>
<tr>
<th>Dye</th>
<th>Rhodamine 590</th>
<th>DCM</th>
<th>Coumarin 450</th>
<th>LDS 698</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Pumping wavelength</strong></td>
<td>532 nm</td>
<td>532 nm</td>
<td>355 nm</td>
<td>532 nm</td>
</tr>
<tr>
<td><strong>Fluorescence peak</strong></td>
<td>596 nm</td>
<td>627 nm</td>
<td>448 nm</td>
<td>692 nm</td>
</tr>
<tr>
<td><strong>Max efficiency</strong></td>
<td>26 %</td>
<td>27 %</td>
<td>15 %</td>
<td>20 %</td>
</tr>
<tr>
<td><strong>Target species</strong></td>
<td>OH</td>
<td>CH</td>
<td>NO</td>
<td>NO</td>
</tr>
<tr>
<td><strong>Transition</strong></td>
<td>$A^2\Sigma^+-X^2\Pi(1,0)$</td>
<td>$C^2\Sigma^+-X^2\Pi(0,0)$</td>
<td>$A^2\Sigma^+-X^2\Pi(0,0)$</td>
<td>$A^2\Sigma^+-X^2\Pi(0,0)$</td>
</tr>
<tr>
<td><strong>Dye laser wavelength</strong></td>
<td>586 nm</td>
<td>628 nm</td>
<td>452 nm</td>
<td>678 nm</td>
</tr>
<tr>
<td><strong>Excitation wavelength</strong></td>
<td>283 nm</td>
<td>314 nm</td>
<td>226 nm</td>
<td>226 nm</td>
</tr>
<tr>
<td><strong>Section</strong></td>
<td>4.1 5.1 5.2 5.3</td>
<td>7.1 7.2</td>
<td>6.1</td>
<td>6.1</td>
</tr>
</tbody>
</table>
3.1.3 Harmonic Generation

Harmonic generation is a nonlinear optical effect that produces radiation at integral multiples of the fundamental (initial) radiation frequency. This is how 532-nm light is produced from an Nd:YAG that operates with a fundamental wavelength of 1064 nm. Notice that wavelength and frequency are inversely related so that the frequency-doubling of the YAG laser results in halving of the wavelength. Using harmonic generation, the output from a laser with desirable parameters but unsuitable wavelength can be frequency doubled, tripled, etc. to form an appropriate beam. For example, an Nd:YAG can be used for the infrared fundamental radiation ($\lambda = 1064$ nm), or frequency doubled to produce green light ($\lambda = 532$ nm), tripled to generate UV ($\lambda = 355$ nm). A fourth harmonic is even used for deeper UV light ($\lambda = 266$ nm). Dye lasers are often frequency doubled too. In Table 3.2, the excitation wavelength is half of the dye laser operating wavelength. This is because the dye laser beam is frequency-doubled, except in the case of LDS 678 which was frequency-tripled.

The strength of harmonic generation is a material property described by non-linear coefficients. Efficient frequency-doubling, or second-harmonic generation (SHG), requires a high second-order non-linear coefficient, $\chi^{(2)}$, and the intensity of a second-harmonic scales with the square of the fundamental intensity. Suitable non-linear crystals for harmonic generation can also be birefringent, where index of refraction depends on the crystal axis. Frequency-doubling is a result of constructive interference and requires the phase velocity of the fundamental and second harmonic be equal. This occurs when the index of refraction at the fundamental frequency is equal to the index of refraction at twice the frequency along a perpendicular axis. Adjust the crystal’s orientation can facilitate this phase-matching. Usually, the crystal is rotated by the phase matching angle, $\alpha_{pm}$, about an axis perpendicular to the two optical axes.
Figure 3.3 illustrates critical phase matching. The crystal index of refraction is also temperature dependent and therefore the phase matching angle is a function of temperature. This can be used as an alternative tuning method, where the crystal temperature is controlled to shift $\alpha_{pm}$ to the crystals fixed angle. Three types of nonlinear crystals were used in this study:

- beta barium borate ($\beta$-BaB$_2$O$_4$ or BBO),
- lithium triborate (LiB$_3$O$_5$ or LBO),
- and potassium dihydrogen phosphate (KH$_2$PO$_4$ or KDP)
Figure 3.3  A plot showing the index of refraction as a function of the angle between the light path and the optical axis for a birefringent material. There is a function plotted for each of the four combinations of frequency and perpendicular axis. The index of refraction in the ordinary direction $n_o$ is constant, and so these functions are circles. The index of refraction in the extraordinary direction $n_e$ is not constant, yielding ellipses instead. The phase matching angle occurs at the intersection of $n_o$ at frequency $v$, and $n_e$ at twice the frequency.
3.2 Imaging

3.2.1 Cameras

In this work, data acquisition utilizes cameras configured to record the desired light signal from fluorescence, scattering, or emission, while minimizing collection of other light (noise). Two camera architectures are used, CCD and CMOS. Both types capture images by converting light into electrical energy through the photoelectric effect. CCD cameras have been used for low frame rate applications (10-Hz PLIF) while CMOS cameras have been used for high-speed applications (10-kHz PLIF) in this study.

A. CCD camera

A charge-coupled detector (CCD) is an imaging device that measures photon-induced charge over an array of metal-oxide-semiconductor (MOS) detectors called pixels. The MOS detectors are produced by layering photosensitive high purity silicon onto silicon dioxide and a polysilicon electronic gate structure. The other side of the photosensitive silicon is attach to a thick sheet of a low resistivity silicon substrate [33]. When the MOS detector absorbs a photon with energy exceeding the bad-gap of silicon and electron-hole pair can be created. The gate structure serves to create potential wells that are very effective at storing the generated electrons. The stored charges are read out by transferring charge from pixel to pixel by manipulating the gate potentials. Usually, an entire row of charges will be transferred to an adjacent row. The last row is a serial register that quickly shifts the charges along the row of pixels to be one at a time and digitized. The charge transfer efficiency is incredibly high, up to 0.9999995 have been reported [73]. The framing rate is limited by the time it takes to read the charges out for each pixel. This delay has been reduced by the introduction of the interline transfer CCD, has rows of masked pixels distributed in between each row of normal pixels. The light induced charged can be
transferred from the unmasked pixel rows to the adjacent row of masked pixels, allowing the unmasked pixels to gather signal from a new image at a higher framing rate [74]. The disadvantage of interline transfer is the reduction in unmasked pixel area, reducing the quantum efficiency of the camera. An array of microlenses can improve quantum efficiency by directing light to the unmasked regions, increasing the fill factor.

The background noise for CCDs is very low, and because the dark current is a thermal effect, the noise can be further reduced by cooling the detector chip. For scientific applications, CCDs are commonly cooled to -20°C to -30°C. Some CCD cameras are capable of hardware binning, wherein the signal from multiple pixels is combined into one larger pixel. This increases the signal-to-noise ratio (SNR) at the expense of decrease spatial resolution. Applications sometimes call for very short exposure to capture a fast event or to bias the signal collection towards a short lived emission. As a solution, image intensifiers can be integrated into a camera to provide gating as well as amplification. Section 3.2.2 Image Intensifiers contains more information on these intensified charge-coupled detectors (ICCD).

**B. CMOS Camera**

A complementary metal-oxide semiconductor (CMOS) is another camera technology. As with CCD cameras, CMOS sensors convert photons into electrical energy by the photoelectric effect. The major difference is that each the conversion from electrical charge into voltage occurs at each CMOS pixel site, whereas a CCD converts charge to voltage at a signal output node. This allows random access to each CMOS pixel yielding higher framerates, but requires a metal grid that reduces fill factor. Additional processing can be performed at each pixel site, yielding greater functionality on-chip [75].
Initial CMOS sensors were composed of passive pixels. Each pixels site included one transistor and provided no amplification. This allowed for good fill factor but the lack of amplification at the pixel site resulted in poor signal-to-noise performance. Modern CMOS sensors contain active pixels, with multiple transistors and amplification in each pixel. Small cameras, such as in phones, web cameras, machine vision, and automotive applications, are almost entirely CMOS sensors because of the low voltage requirement and ability to pack greater functionality on-chip while maintain low cost fabrication methods. The non-uniformity of the sensor, due to the variation in noise and performance of each pixel, is small enough to not be an issue in these high volume consumer applications. In the scientific community, CCDs have remained on top for applications requiring the best possible signal-to-noise ratio and uniformity while CMOS sensors provide electronic shuttering and the highest continuous pixel readout rates and therefore frame rates. This state of the market may change as CMOS technologies improve. In 2009, the launch of the scientific CMOS (sCMOS) sensor was announced as a collaborative project by three large manufacturers of scientific imaging equipment (Andor, Fairchild, and PCO) [76]. These sensors utilize pinned-photodiode pixels with a five transistors (5T) design, facilitating global shuttering, anti-blooming drains, correlated double sampling and sub-microsecond readout times. The sCMOS sensor offers greater resolution, dynamic range, frame rate, and lower read noise than the interline CCD while maintaining a similar quantum efficiency. For the very lowest light imaging, where resolution, field of view, dynamic range, and frame rate can all be sacrificed in pursuit of the lowest read noise, the electron multiplying CCD (EMCCD) is the best sensor.

### 3.2.2 Image Intensifiers

An image intensifier is an electro-optical device that converts a signal from incoming photons into free electrons and back to photons. During this process, the signal may be amplified and
changed in wavelength. Image intensifiers are commonly able to be gated, allowing for very short exposure times. Image intensifiers may be packaged into a standalone unit known as Intensified Relay Optics (IRO). These intensifiers include a method for attaching a lens on the input side and optics at the exit to focus the image onto an external camera. Other intensifiers are integrated into a camera body, resulting in an intensified camera. Both systems use the same method of signal conversion and amplification.

There are three primary components to an image intensifier: a photocathode coated input window, a micro channel plate, and a phosphor coated output window. Figure 3.4 demonstrates how an image is amplified by these components. A camera lens focuses an image onto the input window. The photocathode emits an electron when struck by a photon, due to the photoelectric effects. This does not occur for every photon and the likelihood of a photoelectron (an electron generated by a photon) is described by a percentage called the quantum efficiency. When in operation, the photocathode has a negative charge while the front face of the multi-channel plate (MCP) is grounded. This voltage difference drives the photoelectrons from the photocathode and to nearby MCP. The MCP rear face is held at a positive voltage, so that there is a voltage potential in the same direction, from lower to higher voltage, as the photocathode. Each channel of the MCP acts as a photomultiplier. A photoelectron passing through a channel collides with the channel walls, which produce additional, secondary photoelectrons are produced and also collide with the channel walls to produce still more photoelectrons. The voltage across the MCP determines the gain, and the amplification can be on the order of 1000 [77].
Figure 3.4 A diagram depicted the processes in an image intensifier. The drawing is showing just a single pore in the MCP. The entire MCP is circular and disk-like, with many pores pass through it.

The photoelectrons exiting the MCP are accelerated towards a phosphor screen held at a still higher voltage. When a photoelectron impinges on the phosphor, photons are emitted. The photons can then be imaged using a camera. Note, while these photons correlate with incoming photons spatially, their wavelength is different, determined by the properties of the phosphor. This can be used to allow a camera to operate using light that is outside the limits of its detector, such as infrared or ultraviolet light. The intensifier can be gated by holding the photocathode at a positive voltage, causing the MCP to reject photoelectrons, and then applying a pulse to pull it to a negative voltage, causing the MCP to accelerate photoelectrons. The gate duration can be as short as tens of nanoseconds. In the studies presented here, three types of image intensifiers have been used: a camera integrated intensifier, a single-stage IRO, and a two-stage IRO. The intensified camera contains a fiber optic coupler between the image intensifier and camera detector, while the IROs use a lens coupling. The two-stage IRO contains a second photocathode
and phosphor pair to increase the gain. Figure 3.5, 3.6, and 3.7 illustrate these configurations and Table 3.3 details the specifications.

**Figure 3.5** A CCD camera with an integrated image intensifier, commonly referred to as an intensified CCD, or ICCD. This is representative of the PI-MAX (Roper Scientific).
Figure 3.6 A single-stage image intensifier and optics for coupling to a camera, packaged together as integrated relay optics, or IRO. This is representative of the Invisible Vision UVi (Hadland Imaging).

Figure 3.7 A two-stage image intensifier and optics for coupling to a camera, packaged together as integrated relay optics, or IRO. The second stage increases the intensifier gain for high-speed operation. This is representative of the HS-IRO (LaVision).
Table 3.3 The image intensifiers used in this work.

<table>
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<th>Model</th>
<th>Invisible Vision UVi</th>
<th>HS-IRO</th>
<th>PI-MAX 3 1024i</th>
<th>PI-MAX GEN3</th>
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<td>LaVision</td>
<td>Roper Scientific</td>
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<td>Type</td>
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<td>5.1 5.2 5.3 7.2</td>
<td>4.1 7.1 4.1</td>
<td></td>
</tr>
</tbody>
</table>

3.2.3 Other Detectors

A. Photodetectors

Photodiodes are an easy to use tool for temporal alignment of laser pulses and intensifier gating. In this work, fast photodiodes as are used because of their short rise times (~1 ns). This fast response is needed because the timescales for laser excitation and fluorescence collection are tens of nanoseconds. A quartz window can be inserted into the laser beam path “pickoff” a portion of the laser beam and directed towards the photodiode. This sampled energy is a small fraction of the total, and can be reduced further by using a Brewster’s angle in the plane of polarization. The signal can be displayed on an oscilloscope, conveying timing, energy, and laser pulse profile details. The DET10A (Thorlabs) is the most common high-speed photodetector used throughout these experiments. It includes a fast PIN photodiode and an internal bias battery. Figure 3.13 contains a diagram of the operating circuit.
A PMT is comprised of an evacuated tube housing a photocathode, dynodes, and an anode. For detection of ultraviolet (UV) light, the tube must be a material with good UV transmission. Photons entering the PMT hit the photocathode, causing electrons to be ejected due to the photoelectric effect. These emitted electrons are directed by an electric field to a dynode. Each impinging primary electron causes a number of secondary electrons to be emitted from the dynode. The secondary electrons are directed towards another dynode where still more electrons are released. on the dynode produces collide with the charged walls, producing electrons. This process continues from dynode to dynode until the electrons are collected by the anode, producing a current when a measuring instrument is connected between the anode and grounded photocathode. The gain can be controlled by adjusting the voltage differential across the PMT. A photomultiplier tubes (PMT) can amplify signals at a gain of $10^3$ to $10^8$ with high signal-to-noise ratio. Figure 3.9 diagrams a generic side-on type PMT.
In this work, we use a Hamamatsu R1784 PMT attached to a 0.1 m monochromator (Jobin Yvon). This side-on PMT has a bialkali photocathode and a spectral response range of 185 nm to 680 nm. With five dynodes, the tube has a high pulse linearity and fast rise time (1 ns) at the expense of high gain. Optics focus fluorescence from a small reference burner into a monochromator, and a select band of wavelengths can reach the monochromator exit based on grating position and slit width. This configuration allows for simple excitation scan and monitoring of fluorescence power throughout the experiment.
3.3 Plasma Torches

Two plasma torches have been used in this study: include a microwave plasma system designed for the study of plasma enhanced combustion, and a direct-current (DC) transient-arc plasmatron that produces radical rich turbulent flames.

3.3.1 Microwave Plasma Torch

A novel microwave waveguide plasma system has been developed to study plasma enhanced combustion with various flame types and geometries using laser diagnostics. The microwave plasma source (MPS) was designed to produce plasma discharges spatially coincident with a flame, termed direct coupling, that are accessible for optical and laser diagnostics. The plasma system allows for complete access of the plasma enhanced flame for laser and optical diagnostics at combustion power levels up to about 1 kW. Microwave radiation can be generated in excess of 1 kW but also as low as 30 W. Figure 3.10 shows a plot of operating power ratios, expressed as the absorbed plasma power to combustion power, ranging from 5 to 50 percent for the various conditions.
Figure 3.10 Plot of plasma to combustion power ratio the torch operates at. The shaded region shows the total operable region. Flow rate is the total flow rate for a stoichiometric premixed CH$_4$-air flame.

This system can be compared with other modern microwave devices in the field. A waveguide based MPS has been used to demonstrate enhancement of flame speed by inserting a flat-flame burner into a high-Q microwave cavity power such that the flame is sits within a sub-critical high electric field [24]. Using a pulsed magnetron has been shown to improve energetic efficiency [78]. These constructions serve to maximize efficiency, but can complicate diagnostics due to restricted optical access to the discharge and/or flame. Other devices use a coaxial design with differing electrode lengths, so as to improve the efficiency of the field transition to a surface wave [79]. The present system can be described as a modification to the well-studied TIAGO (Torche à Injection Axiale sur Guide d'Ondes) MPS, which has been shown to produce atmospheric plasmas accessible for diagnostics [80]. The MPS presented in this study is an advancement because it is able to produce optically accessible discharges that allow for study of a direct coupled plasma enhanced flame with the flexibility for a variety of conditions to further
the study and application of plasma assisted combustion. While the MPS efficiency is not poor, capable of incident to reflected power ratios of better than 10:1, optimization of this system is secondary to the design parameters of discharge accessibility and flexibility.

This system has been developed to produce a direct coupled plasma enhanced flame accessible to optical and laser diagnostics. The torch would likely be more efficient with a truncated inner electrode, where the outer electrode (nozzle body) extended further than the inner solid electrode, but a heavy interest in complete access to the plasma ignition and coupling point directed the decision to use a “flush” configuration, where the electrodes end at the same length. Additionally, importance was placed on the flexibility of the system to allow for study of various flame types and geometries, flow rates, gas flow compositions, etc.

Figure 3.11 shows a schematic diagram of the MPS. The microwave electromagnetic radiation is generated by a research grade 2.45 GHz magnetron (National Electronics) controlled by a continuous power generator (Alter Power Systems) with a maximum power of 2 kW. The microwave transmission inside the WR284 waveguide is in the TE_{10} mode. A directional coupler measures the power of the wave exiting the magnetron head. An isolator, consisting of a circulator and water-cooled dummy load, allows for measurement of reflected microwave power (National Electronics). The plasma applicator nozzle facilitates the plasma flame coupling, functioning as a coaxial waveguide and torch (Amarante Technologies). The waveguide is tuned through use of a three stub tuner (National Electronics) and sliding short (Gerling).
Figure 3.11 Diagram of microwave waveguide plasma system.

Figure 3.12 illustrates the plasma applicator and nozzle, which also acts as the torch for combustion. The nozzle has a solid electrode that protrudes inside the WR284 waveguide. Microwave energy travels through the nozzle, acting as a coaxial guide, in the TEM mode. The internal geometry of the nozzle is specially optimized for plasma ignition by delivering a high electric field at the nozzle tip. A plasma discharge is initiated by adjusting the standing wave’s maximum E-field position inside the waveguide using the sliding short. As the standing wave moves inside the waveguide, the E-field at the nozzle tip also varies. When the E-field reaches the breakdown threshold, air around the electrode tip will be ionized generating a plasma discharge that sits entirely above the surface of the nozzle and is accessible to direct optical measurements. Initially, the solid electrode plasma nozzle experience occasional arcing within the nozzle body. Increasing the flow rate through the nozzle corrected this problem, but limited the interaction between reactant flow and plasma volume. A better solution was found by incorporating a quartz tube as a dielectric barrier. The quartz tube outer diameter is close enough
to the inner diameter of the nozzle that the tube can be wrapped in single layer of PTFE tape (as is used for tapered pipe fittings) and pressed into place. The inner diameter of the quartz tube is 5 mm, allowing sufficient area for flow around the ~0.92 mm diameter electrode. Material choice of PTFE and quartz in the design is important to maintain the waveguide structure and the microwave waveguide can be tuned to account for deviation in impedance.
Figure 3.12  Diagram of the plasma applicator nozzle (left), photograph of the plasma applicator nozzle in operation with a plasma coupled CH₄-air premixed flame (right).

### 3.3.2 Direct-Current Transient-Arc Plasmatron

The discharge system used in this study to generate hot NO is a transient-arc direct-current (DC) plasmatron [20, 81, 82] from Applied Plasma Technologies; a schematic is shown in Figure 3.13. This plasma torch operates on a repetitive glow-to-spark transition mode, offering the advantages of a thermal plasmatron with low average power output, low average temperatures (600–1300 K) [83], and high electron density. The torch operates primarily in the glow mode where high voltage (~10 kV) is applied between the cathode (inner electrode) and the surrounding anode.
During the glow-to-spark transition, a short-duration spark (~100 ns) propagates from the cathode towards the torch tip. As the overall temperature is low, the electrode erosion is significantly less than with traditional DC torches, and the torch can produce stable operating conditions without internal cooling. The plasmatron exit has an inner diameter of 5 mm. Quantitative measurements of NO have been performed previously at 10 Hz acquisition frequency using this plasmatron, with concentrations reported as 5000 to 15000 ppm in an air-plasma and 500 to 3500 ppm in a plasma-assisted flame [83, 84].

![Diagram of DC plasma torch and photographs of air-plasma and plasma-assisted flame](image)

**Figure 3.13** Diagram of DC plasma torch (left), and photographs of air-plasma (top right) and plasma-assisted flame (bottom right).

There are two gas inlets in this discharge system: Feed 1 guides gas flow directly to the arc chamber, and Feed 2 provides coflow around the main exit of the torch. An air-plasma is
produced by flowing compressed dry air into Feed 1 at 6 slpm, yielding an exit velocity of 5 m/s, and applying a voltage potential between the electrodes. For plasma-assisted combustion, CH₄ is added to the feedstock gas (Feed 1) at the proper rate to yield a stoichiometric mixture at a total flow rate of 6 slpm, and the plasmatron ignites and sustains a flame. Air is directed through Feed 2 at 10 slpm, acting as a coflow for the cold flow case. It is noted that the flowrates used in this study are too high for unassisted flame stabilization.
3.4 Wind Tunnel

The supersonic experiments presented here (see section 5.1 10-kHz OH PLIF in a Supersonic Wind Tunnel Cavity Flameholder) were performed in the direct-connect supersonic research facility at Wright-Patterson Air Force Base (Research Cell 19). Air flow through compressors and a gas-fired heat exchanger produce a high-enthalpy “hot line” at up to 17 lbm/s at 750 psig and 1660°R (5.18 MPa and 922 K at 7.7 kg/s), and a “cold line” at up to 17 lbm/s at 750 psig and ambient temperatures [85]. The hot and cold lines merge at a mixing station and are used to create the desired conditions. The mixed air flows through an insulated expansion loop to a supply manifold. Three branches of the manifold supply air to the research cell, while the two branches exit out of the roof, serving as a vent line and a pressure relief line. The facility is capable of continuous flow with a range of Mach number, and peak stagnation conditions of 400 psig and 1660°R at a peak flowrate of 34 lbm/s (2.76 MPa and 922 K at 15.4 kg/s) [85]. Back pressure can be manipulated remotely by a control valve in the facility exhaust section. Air delivered to the test cell is distributed as it passes through a rearward facing perforated cone in the expansion section. Then, the flow passes through the settling chamber, conditioned by an array of mesh screen and honeycomb. A transition region connects the axisymmetric settling chamber to the planar nozzle section. Nozzles are two-dimensional and designed by a computational code using the method of characteristics [86]. The supersonic air travels through an isolator section and into the test section. Air exiting the test section is cooled by water-spray and decelerates through a diffuser. Greater details on the design and fabrication can be found in ref [87] and calibration in ref [85] by Gruber et al. The supersonic research facility has been used a variety of studies, including: ignition by plasma [88] and pulse detonation [89]; enhanced mixing and flame holding by struts [90], pulse detonator [91], and cavity [92, 93]; flow field velocimetry [94]; and inlet distortion [95-97].
For this study, continuous air flow is conditioned by the settling chamber, accelerated through an asymmetric nozzle to Mach 2, and then passed through a constant-area isolator section measuring 50.8-mm high and 152.4-mm wide before entering the test section where the floor diverges at 2.5°. The recessed wall cavity is 76.2 mm downstream from the start of the test section and measures 16.5-mm deep and 66-mm long (to the midpoint of the closeout ramp), while spanning the tunnel width of 152.4 mm. The upstream rearward-facing step is a 90° corner while the closeout ramp is at 22.5° relative to the duct floor. Ethylene (C₂H₄, chemically pure grade) is injected through a spanwise row of 11 fuel injectors in the ramp, such that the fuel jets are parallel to the floor and opposing the freestream flow direction, following the recirculation pattern within the cavity. Additional fuel can be injected into the main tunnel flow upstream of the cavity through a 25-mm wide floor slot in the spanwise center of the floor. Fused-silica windows allow for optical access from both sides and the top of the test section. The test section is optically accessible from three sides, with large UV-grade fused silica windows on each side spanwise that extend well beyond the cavity length streamwise and beyond the test section and cavity height. A window of a width about half of the spanwise dimension can be placed in the top wall of the test section, used in this work to pass the PLIF laser sheet flameholding cavity. During this experiment, the respective total temperature and pressure were 611 K and 483 kPa.
4.1 Microwave Plasma Torch 10 Hz diagnostics

A tunable microwave waveguide is used to initiate and enhance combustion by coupling an atmospheric plasma discharge to a premixed methane/air flame. The absorbed microwave power ranges from 60 to 150 W, generated from a continuous source operating at 2.45 GHz, whereas combustion power ranges from 200 to 1000 W. OH radical number densities were measured using planar laser-induced fluorescence (PLIF) and temperatures were measured using Rayleigh scattering thermometry for various flow rates, equivalence ratios, and power levels. Increases in reaction volume, OH density, and temperature were observed as power increased.

Premixed and non-premixed flames are studied, using both solid and hollow inner conductors in the plasma applicator nozzle. Plasma power is controlled by adjusting the reflected microwave power, measured at a dummy load attached to a circulator. Maximum OH radical number densities were quantified as approximately \((3-5) \times 10^{16} \text{ cm}^{-3}\) for plasma powers around 100 W, with small variation between configurations. The maximum temperatures occurred in the non-premixed flame, where the plasma is generated in air, reaching values of 3500 K. Temperatures are lower, peaking at 2000 K, when the plasma is generated at the air-fuel boundary or the air-premixed boundary through use of the hollow inner conductor. Additional parameters are adjusted, including flow rates, power level, equivalence ratio, and the effects are discussed. Non-premixed configurations are ill-suited for flame enhancement, whereas a premixed flow through the hollow electrode best demonstrates non-thermal plasma assisted combustion.

Temperature in the plasma coupled premixed flame, ranging from 2000-3000 K, increased by up to 40 percent as the absorbed microwave power was increased from 60 to 130 W. Air-only
Plasma discharges exhibited a much greater temperature increase, up to 190 percent. The power associated with the measured temperature increases varied greatly with flow and input power, but are typically 3 to 4 times greater in the air only plasma compared to the flame coupled plasma, demonstrating a greater degree of non-thermal mechanisms present in plasma enhanced flame discharge.

4.1.1 Introduction

Electromagnetically enhanced oxidation holds great potential to be the foundation of an improved energy infrastructure by facilitating highly efficient thermal energy conversion through plasma assisted combustion [5, 6]. There are many benefits to applying electromagnetic energy to a combustion reaction, including faster and more intense chemical energy conversion, increased flame stability over a broad range of conditions, reduction of pollution, improved fuel efficiency, and more reliable and rapid ignition [8-10, 12, 13, 98]. Plasma assisted combustion is particularly well suited for use in challenging conditions, as present in hypersonic flight, where complete and stable combustion is exceedingly difficult using traditional methods [12, 13].

The efficiency of a plasma assisted combustion system, as a measure of the value of energetic enhancements versus its energy use, is critical in the assessment of such a system for practical implementations. In previous work, the use of a highly efficient direct coupling method was investigated, where a plasma is generated spatially coincident with the flame reaction zone [99], enabling plasma induced flame enhancement at very low power levels. The plasma and flame complement each other, as elevated temperature and reactivity of the plasma discharge initiates and enhances combustion, and the free electrons and heat release in the flame lower the power needed to sustain the plasma and increase the level of ionization. The present study furthers the demonstration and understanding of direct coupling by extending its application to more realistic
flame geometries and higher combustion powers. The study analyzes the physicochemical effects of direct coupling through imaging of spatially resolved hydroxyl (OH) concentrations and temperature fields using planar laser-induced fluorescence and Rayleigh scattering, respectively.

4.1.2 Experimental Setup

A. Microwave Waveguide Plasma Applicator

The torch nozzle allows for gas flow into the plasma discharge. To produce a premixed flame, a mixture of fuel and air enters into the side of the aluminum nozzle through inlet (a), and exits into the plasma discharge at the tip of the electrode (b). A Teflon insert prevents the flow from entering into the microwave cavity at the bottom off the nozzle, while allowing microwave energy to travel upwards (c). This configuration produces a direct-coupled plasma assisted combustion discharge. A coflow adaptor allows for an annular stream of gas around the nozzle but was not needed here. In data presented here, the reactants are methane and air. Flow rates are measured in SLPM (standard liters per minute), following the IUPAC standard temperature and pressure as 273.15 K and 1 standard atm.

The plasma applicator nozzle can be configured to produce premixed and non-premixed flames, while using a solid electrode or a hollow grounded electrode, allowing for examination of various flame structures. Four configurations are presented in this study: 1) a solid electrode with premixed flame, 2) a solid electrode with non-premixed flame 3) a hollow electrode with non-premixed flame, and 4) a hollow electrode with premixed flame. As illustrated in Figure 4.1, an additional gas flow path is added to produce the non-premixed flame with the solid electrode, whereas the same geometry is used to produce both flame types with the hollow electrode through adjustment of the gas flow inside the electrode. A Teflon insert holds the solid electrode.
in position while preventing the nozzle from entering the waveguide. Furthermore, to prevent arcing, a short quartz tube is inserted flush with the nozzle tip. The MW power level is defined as the difference between measured incident and reflected power. A desired power level was achieved by adjusting the tuning components to achieve a reflected power corresponding to the desired absorbed power level. The incident power remained fixed at about 360 W, the lowest possible value for the power source. Losses were assumed to be minimal and not accounted for, as the power level values are used for relative comparison and not absolute calculations in the study.

![Diagram of microwave plasma waveguide system nozzle configurations](image)

**Figure 4.1** Diagram of microwave plasma waveguide system nozzle configurations: solid electrode premixed, solid electrode non-premixed, hollow electrode premixed or non-premixed (bottom, left to right).

**B. Laser Diagnostics**

OH was excited using the $Q_1(8)$ transition of the $A^2Σ^+-X^2Π$ (1,0) band, which requires narrowband UV light near 283 nm [3]. The measurements were conducted using a dye laser (Lumonics Hyperdye HD-300) with an output of 7 ns pulses at 566 nm, which was subsequently
frequency doubled through an Inrad Autotracker (ATIII) to the desired excitation wavelength of 283 nm. The laser is pulsed at 10 Hz and has a spectral line width of about 0.1 cm$^{-1}$ at 283 nm. The pulse energy was recorded digitally using a fast photodiode and an oscilloscope and attenuated (using a combination of a polarizing beam splitting and a half-wave plate) to ensure operation within the linear fluorescence regime. The laser beam was expanded into a sheet and the fluorescence signal was collected at 90° using an intensified CCD camera (Roper Scientific Superblue PIMAX) fitted with a Cerco 45-mm focal-length, f/1.8 lens and a high-transmission (>80% at 310 nm) bandpass filter (Custom fabrication – Laser Components GmbH). A PMT coupled to a small spectrometer with collection optics focused on a flat flame burner was used to identify excitation spectra and monitor the laser wavelength. A Hencken burner was used to correlate the signal intensity with the OH number density using previously performed absorption measurements.

Rayleigh scattering thermometry was conducted using the frequency doubled output of an Nd:YAG laser (λ=532 nm). Scattering measurements were used here, rather than other thermometry methods that are based on assumptions of the population of quantum states, due to the non-equilibrium nature of the plasma discharge. Scattering was collected through a bandpass filter centered at 532 nm with a 10 nm FWHM using a second intensified CCD camera (Roper Scientific Gen III PIMAX) fitted with a Nikon 58-mm focal length f/1.2 Noct-Nikkor lens. Black curtains enclosed the test section to limit background scattering, and sufficient signal quality was achieved so that Filtered Rayleigh scattering approaches were not required. The collected scattering is a measure of number density from which a planar temperature profile can be obtained utilizing the Ideal Gas Law. By normalizing the images by an image of Rayleigh scattering in air with no discharge, the laser intensity is eliminated from the calculation. The temperature is given by
\[ T_d = \frac{\sigma_d I_a}{\sigma_a I_d} \cdot T \]  

where \( \sigma_d \) and \( \sigma_a \) are the respective Rayleigh scattering cross sections of the discharge gas and air. \( I_a \) is the reference scattering intensity in air at temperature \( T_a \) and \( I_d \) is the scattering intensity from the plasma discharge. For premixed combustion cases with an equivalence ratio near \( \phi = 1 \), gas composition in the was assumed to be the stoichiometric products of CH\(_4\)-air, which limits error due to composition assumptions to less than 10\%, as nitrogen is still the primary constituent. For the very rich premixed flames, this assumption leads to an underestimation of temperature in the reactants region. The magnitude of the error is proportional with the fuel fraction in the reactants. No correction has been made because the composition is unknown and cannot be reasonably estimated or modeled due to the turbulent nature of the flame. This will be further discussed with the presentation of any results of affected cases. For air plasmas, composition of dry air was used for the cross sections. To reduce Mie scattering, air and reactant flows were filtered, the camera was positioned to collect backscattering, and averaged images were calculated as the median of 200 images.

### 4.1.3 Quantitative Methods

Quantitative planar laser measurements are challenging in reactive flows, and even more so in a turbulent flame. Both OH PLIF measurements and Rayleigh scattering thermometry measurements are susceptible to error and uncertainty, resulting from assumptions of gas composition. PLIF measurements additionally suffer from estimations of temperature. Here, we will discuss these uncertainties and their effect, beginning with OH PLIF.
A. Absorption Measurement

The OH radical concentrations were quantified using an absorption measurement in a laminar flame. This calibration measurement itself involves assumptions and signal corrections as discussed by Yin, Carter, and Lempert in ref [100]. In their work, a 25-mm square Hencken burner (Technologies for Research, model RD1X1) was used to make an OH absorption measurement in a premixed, laminar flame. The near-stoichiometric reactant mixture ($\phi \approx 0.95$) of CH$_4$ and air at a total flow rate of 10.6 SLPM. The CH$_4$ was of 99% purity and the compressed air was filtered to remove oils and water vapor. Mass flow controllers (Tylan) metered the gas flow rates and were calibrated by a Bios Drycal unit. A coflow of N2 issued from the burner to reduce instability. A frequency doubled, tunable dye laser was pumped by an injection-seeded Nd:YAG to produce a 309-nm wavelength beam with a pulse width of 8 ns and estimated linewidth of $\sim 0.2$ cm$^{-1}$. The pulse energy before and after the laser beam passed through the flame was recorded using a joulemeter (Molelectron) attached to an integrating sphere. An ICCD camera (Roper Scientific PIMAX) placed perpendicular to the beam path was synchronized to the laser beam and recorded LIF.

The absorption of the laser beam by OH radicals in the flame was measured by tuning the dye laser to OH $A^2E^+\leftarrow X^2\Pi$ (0,0) transitions. The absolute OH number density $n_{OH}$ can be found using the equation

$$Absorbance = \int_{-\Delta v}^{+\Delta v} \left[1 - \exp\left(-h\nu_0 g_a(v, \nu_0, T, P) f_B n_{OH} L_{abs}(x) \frac{B_{12}}{c}\right)\right] dv,$$  \hspace{1cm} (4.2)

where $h$ is the Plank constant, $\nu_0$ is the center of the transition, $g_a$ is the normalized absorption lineshape (a function of temperature, pressure, transition center, and incident frequency), $f_B$ is the Boltzmann fraction of the OH molecules in the absorbing state, $B_{12}$ is the Einstein transition
probability, and \( c \) is the speed of light. The absorption path length \( L_{\text{abs}} \) was determined from the simultaneous LIF measurements using

\[
L_{\text{abs}}(x) = \frac{1}{S_f(x = a) \int_{-\Delta x}^{+\Delta x} S_f(x) dx},
\]

(4.3)

where \( x \) is the distance along the laser beam path with an origin at the over the burner center, and \( a \) is the calibration location.

The normalized lineshape \( g_a \) is a convolution of the Doppler and collisional broadening lineshapes. The collisional lineshape was calculated using the experimental results from Rea et al. in ref [101, 102]. The temperature was assumed to be 2150 K for lineshape and Boltzmann fraction calculations. The transitions measured are relatively temperature insensitive over the range of 1500 to 2000 K. With all sources of uncertainty considered (\( T, L_{\text{abs}}, b_{12}, \) collisional broadening, etc.), Yin et al. report an absolute OH number density at the point 25 mm over the center of the burner as 
\( n_{\text{OH}} = (0.93 \pm 0.05) \times 10^{16} \text{ cm}^{-3} \) [100].

**B. Quantitative OH PLIF**

The time-integrated fluorescence signal \( S_f \) (here with units of detector counts) from collection volume \( V_c \) is

\[
S_f = V_c \beta \frac{\Omega}{4\pi} f_b N_{\text{OH,abs}} \hat{F}
\]

(4.4)

\( B \) accounts for the net optical transmission and detector gain, \( \Omega_c \) is the solid angle of detection, and \( n_{\text{OH}} \) is the OH number density. The specific fluorescence \( \hat{F} \) is defined in the linear regime as

\[
\hat{F} \equiv \int_{\Delta t} b_{12} dt \cdot \varphi_f
\]

(4.5)
\( \phi_J \) is the fluorescence yield,

\[
\varphi_J = \frac{\sum_j f_{j,v}^0 \cdot A_{j,J}^{v,J=1} + \sum_j f_{j,v}^{0 \rightarrow 0} \cdot A_{j,J}^{v,J=0} \cdot \sum_j f_{j,v}^{0 \rightarrow 1} \cdot V_{J,v}^{10}}{\sum_j f_{j,v}^{0 \rightarrow 1} \cdot (A_{j,J}^{v,J=1} + Q_{J}^{v,J=1} + V_{J,v}^{10})}
\]  

(4.6)

where \( A_{j,J}^{v,J=1} \) is the sum of the spontaneous emission rate constant for each possible transition for a given upper vibrational and rotational state, \( Q_{j,v}^{J} \) is the J-dependent electronic quenching rate, and \( V_{J,v}^{10} \) is the vibrational energy transfer rate from \( v' \) to \( v'' \). The electronic quenching rate and vibrational energy transfer rates are the most challenging term, with dependence on gas composition and temperature. The time-integrated population fraction \( f_{j,v}^{0 \rightarrow 1} \) is also problematic. For this term, two extreme cases can be analyzed: 1) completely thermal equilibrium in the excited state rotational manifold; 2) no rotational energy transfer, meaning the excited state population maintains its rotational level until fluorescence. The actually distribution is somewhere between these two extremes. Similarly, the quenching and vibrational energy transfer rates can be evaluated at two extreme cases to give upper and lower boundaries. These boundaries, along with uncertainty in the spontaneous emission rates collected from LIFBASE, allow for a calculation of an upper and lower boundary for the fluorescence yield \( \varphi_J \).

In the linear regime, fluorescence signal is directly proportional to the fluorescence yield, and so the signal carries (at least) the same uncertainty. In the experiments here, some of the flows are very rich premixed gas mixtures, with equivalence ratios as great as 8.0. This affects flame temperatures, and of course gas mixture composition. First, a baseline case was considered. Here, CHEMKIN is used to model a stoichiometric CH4-air freely propagating flame. The
fluorescence yield can be from the calculated species mole fractions and temperatures. The quenching and vibrational rates are calculated as the mole weighted average of the gas components using experimentally determined cross sections $\sigma_p$ from [103], where $p$ is the collisional partners, via

$$Q_p \text{ or } V_p = n_0 v_{OH} \sum_p \chi_p \left(1 + \frac{m_{OH}}{m_p}\right)^{1/2} \sigma_p$$  \hspace{1cm} (4.7)

Where $v_{OH} \equiv \left(\frac{8kT}{\pi m_{OH}}\right)^{1/2}$, $n_0$ is the total number density, and $\chi_p$ is the mole fraction of species $p$.

Corrections for electronic quenching and vibrational energy transfer were first investigated using a freely propagating flame computation in CHEMKIN II PREMIX using the USC Mech II kinetic model [104]. From these results, the temperature and gas composition data for major quenching species were used to calculate the variation in fluorescence yield throughout the flame thickness and into the products, defining the starting contour as when temperature first exceeds 700 K. For this range, the variation in fluorescence yield due to electronic quenching and vibrational energy transfer is 25.6%.
4.1.4 Results and Discussion

A. Argon Plasma

It is important to verify that the current configuration of delivering microwave energy for plasma generation is capable of generating a ‘cold-plasma’ with a high degree of thermal non-equilibrium. For demonstration, a non-equilibrium plasma was produced using the solid electrode configuration with pure argon flowing through the nozzle. The discharge was initiated at absorbed power levels of 35 W and flow rates of 56 to 112 SLPM (60 to 125 m/s), much lower power levels and higher flow rates than plasmas of air and fuel. A cold argon plasma can be sustained at temperatures near ambient conditions. After the plasma is initiated, increases in power level quickly produce a temperature increase in and around the plasma. Figure 4.2 graphically illustrates and plots the discharge temperature for various power levels and flow rates. Plotted data are the mean values of a small (1 mm × 0.75 mm) region in the averaged image for the given condition. The ability to generate low power, low temperature plasma at atmospheric pressure has been of interest as a sterilization method, particularly for heat sensitive materials for which traditional sterilization is not suitable [105, 106]. The non-thermal nature of the argon plasma contrasts sharply with measurements of an air-only plasma. When the torch is operated with a flow composition of only dry air, temperatures were measured in excess of 3000 K. Additionally, inclusion of small amounts of oxygen or methane into the argon plasma feedstock immediately raised the plasma temperature.
**Figure 4.2** Images of average temperature [K] of an argon plasma discharge for varying microwave power levels and flow rates (left), plot of temperature [K] versus power [W] for multiple flow rate.

**B. Air-Only Plasma Discharge**

Imaging results are shown in Figure 4.3 as a function of increasing microwave power for just the plasma discharge in air without combustion. All images and photographs are at a flow rate of 15 SLPM. The first row contains instantaneous OH number density images, the second row contains the median temperature images, and the third row contains photographs of the discharge at an exposure of 10 ms. The microwave power level is simply the difference between incident and reflected microwave values from the waveguide. The magnetron output was fixed at its lowest possible value, 360 W, for all discharges, the effective power input into the plasma was adjusted by manipulating the reflected power through tuning. Percent power reflected will then be the difference of the magnetron power (360 W) and reported microwave power, divided by the
magnetron power. A microwave power level ranging from 60 to 150 W corresponds to ~83 to 58%. The waveguide can be tuned to reflect very little power (<10%).

Observation of the image series clearly illustrates an increase in discharge volume and emission, OH number density, and temperature as microwave power increases. Temperature in the air plasma is quite high with a highly focused power deposition. OH production due to the plasma discharge comes solely from the water vapor in the ambient air, reaching densities up to $10^{16}$ cm$^3$ in a laboratory with moderate humidity (40 to 50% relative humidity). This is demonstrated clearly in the OH PLIF images as the center plasma discharge volume is devoid of OH radicals because this is dry, compressed air. Instead, the area above the discharge, where ambient air can

**Figure 4.3** Images of OH number density [$10^{16}$ cm$^{-3}$] (top), averaged temperature [K] (middle), and photographs of discharge (bottom), of an air plasma discharge for varying microwave power levels, increasing left to right (15 SLPM flow rate).
mix with the hot discharge, contains an amount of OH usually associated with levels found in methane/air combustion [98]. It should be noted that OH populations in the Boltzmann fraction are considered to be constant, which is an adequate assumption for temperature range of about 1050 K to 2600 K (<10%). However, additional error is expected at lower temperature regions below 800 K and above 3000 K, where an under-prediction of OH occurs. No adjustment has been made due to lack of simultaneous temperature-OH density data in the turbulent flow field. The bright air discharge in the photographs indicates emissions from all visible wavelengths typically found in this type of plasma (mainly emissions from OH, N₂, N₂*) [107]. The discharge is estimated to be weakly ionized with electron density of about 10^{11} to 10^{12} cm^{-3} with an estimated e-field on the order of the breakdown threshold of air, 10^6 V/m.

Figure 4.4 shows the plotted relationship between OH number density, temperature, and power. Here, the OH number density is defined as the maximum value in an averaged image, calculated as the mean value of each pixel from 200 single images. With no hydrogen source on the dry air flow, there is a lack of radical concentration in the plasma volume. The OH production observed is a product of the water vapor in the ambient air reacting with free electrons produced in the plasma, as well as atomic oxygen. The mechanisms of interests are e+H₂O→e+H+OH [108], and H₂O + O (¹D)→2OH [109].
Figure 4.4 OH number density (top) and temperature (bottom) vs power in the air plasma discharge for varying flow rates.

The OH number density remains relatively stable over a power range of 60 to 90 W. Changes in power level do not significantly affect the OH production chemical pathways. At higher powers, OH number density increases near-linearly with power. OH number density is inversely related to flow rate. This is expected, as a lower flow rate allows for greater power deposition into a given volume of air above the torch. Once again, under-prediction of OH concentrations is expected at very low and very high temperature regions, for which no correction has been made.

Temperature increases as a function of power in a near-linear manner. Like OH number density, the temperature is inversely related to flow rate because of the greater power density at lower flow, which in turn will increase the production of OH radicals from water vapor in the ambient air. The plotted temperature data are the mean values of a high temperature region in the average temperature profiles, calculated as the median of 200 single images. The region was defined as a
0.75 mm by 1 mm area where the mean temperature was greatest. Temperatures range from about 650 K at 60 W and 20 LPM, to 2650 K at 130 W and 5 LPM.

C. Solid Electrode and Premixed Flame

OH number density, temperature, and photographs of a premixed, plasma assisted combustion case are shown in Figure 4.5. For all conditions, the flow rates are significantly fast (a minimum of about 5 m/s) that a flame cannot be naturally stabilized, but must be ‘anchored’ by the plasma discharge. The same trends observed in the air only discharge can be seen in the combustion case; discharge volume, emission, OH number density, and temperature all increase with increasing microwave power. OH number density is significantly higher than in the air discharge due to additional hydrogen atoms available from the hydrocarbon combustion chemistry. The discharge temperature is greater than in the air-only case and the high temperature region is much larger due to the overlap with the expanding flame volume. The photographs show a violet plasma discharge, indicating strong emission in the UV range. The flame emits a blue hue, typical of CH* emission in premixed combustion, accompanied by heat release and increased radical concentration, indicating ignition. Spectrally resolved chemiluminescence of the coupled discharge strongly agrees with that of a flame, confirming combustion.

All data presented are from CH₄/air plasma-enhanced flames. The ignition procedure was to flow the fuel and air, then ignite the flame by increasing effective microwave power to the nozzle tip, through adjustment of the three stub tuner, until breakdown and plasma formation. In all cases the flame is not stable without the plasma discharge; extinguishing the plasma discharge results in flame blow-off.

A premixed flame was ignited and sustained by a microwave plasma discharge using the solid electrode configuration. Microwave power, flame equivalence ratio, and total flow rate were
individually varied as data was collected. A microwave discharge could be observed at an absorbed power of about 70 W, igniting and sustaining a flame for all tested flow rates, from 5 to 20 SLPM (5.5 to 22.1 m/s). After ignition, the plasma discharge is stable down to power levels of 60 W. The flame is not stable without the plasma discharge, and blow-off is observed if the plasma is extinguished by detuning the waveguide.

**Figure 4.5** Images of OH number density [$10^{16}$ cm$^{-3}$] (top), averaged temperature [K] (middle), and photographs (bottom), of the plasma coupled combustion discharge at varying microwave power levels, increasing left to right. Total flow rate is 15 LPM at an equivalence ratio of $\varphi=1$.

OH number density and temperature in the premixed discharge are plotted for a range of absorbed power in Figure 4.6. OH number density for different flow rates remains relatively constant even though the energy per molecule is greater as the flow is reduced. In comparing the OH number density, temperature, and photographic images for different flow rates at constant
power, it is observed that the reaction volume is greater as the flow is lowered, due to the lower momentum of the gas flow, resulting in a dilution of OH radicals. We conclude that the increasing energy per molecule and dilution effects are similar in magnitude, effectively canceling each other. Unlike the air-only discharge, OH number density increases over the entire power range.

For OH concentration calculations, the Boltzmann fraction was considered constant, an adequate assumption for temperatures in the range of 1050 K to 2600 K, as the population varies less than 10% from the calibration flame value. Additional error is expected outside of this range, where an under-prediction of OH occurs due to lower state population. Figure 4.5 displays a series of OH concentration and temperature images with power increasing from 60 to 130 W. As discussed in our previous work, there is a clear increase in OH number density and temperature as the cavity is tuned to increase the absorbed power. Average temperature increased from approximately 2150 to 2650 K as power was increased from 60 to 130 W. Maximum hydroxyl concentration increased from 3 to $4.6 \times 10^{16}$ cm$^{-3}$ for the same power interval. The increase in excited state species produced by plasma chemistry expands the chemical pathways to OH production. For example, oxygen in the form of O($^3P$) and O($^1D$) is produced via $O_2 + e \rightarrow O(^3P) + O(^1D) + e$ [110] and contributes to OH radical concentration through reaction with water vapor $O(^3P) + H_2O \rightarrow OH + OH$, $O(^1D) + H_2O \rightarrow OH + OH$ [111].

In analyzing temperature, we can see that at lower powers and higher flow rates, the temperature (median of 200 single-shot images) is below the adiabatic temperature of the flame (>2200K for CH$_4$-air). This is likely due to incomplete combustion caused by the higher flow velocities, though it is important to note that as median values, these temperatures are not the absolute maximum for a single image. Also it should be noted that for a given flow rate, unlike the air plasma discharge, the increase in temperature as a function of power is relatively small.
appears to be an asymptotic limit to temperature, approached by all flow rate lines where the combustion becomes fully complete. This leads us to speculate that while in the air plasma most of the plasma energy is transferred to the gas as heat, other transfer channels play a significant role when the plasma energy is coupled into a premixed flame where electrons and ions are already present.

Figure 4.6 OH number density (top) and temperature (bottom) vs power in the premix plasma discharge for varying flow rates. Equivalence ratio $\phi=1$. 

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Figure 4.7 shows observed OH number density and temperature as a function of power for various equivalence ratios. Temperature is highest at rich conditions due to greater chemical heat release, while OH number density is maximized in lean conditions due to the abundance of oxygen atoms. Both temperature and OH density increase with an increase in power.

**D. Solid Electrode and Non-Premixed Flame**

A non-premixed flame was produced with the addition of a coflow adaptor. Air flow through the nozzle was a constant 5 SLPM yielding a speed of 5.5 m/s. Fuel flow through the coflow adaptor (annular surrounding the nozzle) varied from 0.5 to 2.0 SLPM (0.4 to 1.4 m/s) As previously described, air is injected in the center of the nozzle while a jet of fuel is injected around the circumference. There are two flame fronts in this configuration; the inner front is caused by fuel entrainment into an air plasma plume whereas the outer front is a typical flame
with the surrounding air. The OH number density along the inner front increases with power and is similar in magnitude to the solid electrode premixed case. The OH density in the outer front is much lower, in the expected range for a diffusion flame. OH number density, temperature, and photographs of a non-premixed, plasma assisted combustion configuration can be seen in Figure 4.8.

![Figure 4.8](image)

**Figure 4.8** Images of OH number density \([10^{16} \text{ cm}^{-3}]\) (top), averaged temperature [K] (middle), and photographs of discharge (bottom), of a solid electrode non-premixed discharge for varying microwave power levels, increasing left to right (1 LPM fuel, 5 LPM air flow rate).

The maximum OH concentration is located within the inner flame front and is largely a function of power, ranging from \((2-5) \times 10^{16} \text{ cm}^{-3}\) for power levels of 60 to 110 W. This is the same range of OH concentration measured previously in a solid electrode premixed configuration. The inner flame height is reduced as power is increased, transitioning from a lifted flame to stable flame at the plasma plume tip as the flame speed increases. The maximum temperature is very high in the plasma discharge volume, in excess of 3500 K at 130 W. Plasma energy has mostly a thermal
effect on the airstream, heating it to a greater degree than the methane-air premixed flame. As expected, maximum temperature is independent of fuel flow rate because the plasma is positioned inside the air jet. An error of up to 10% in temperature is expected due to composition assumptions in this region of only air. However, the area between the inner and outer flame is composed mostly of fuel, and presumably only fuel in the regions near where the fuel is issued. Methane’s scattering cross-section is about twice the cross-section used to calculate temperature (stoichiometric products) and so the fuel only region is actually twice the temperature reported here. Near the flamefront, the fuel rich reactants region is at a higher temperature than calculated. If the data could be corrected with full knowledge of composition for each frame, the expectation is that the high temperature regions would extend slightly more toward the CH₄ only region, reducing the temperature gradient. The contour of the flame front as marked by temperature might shift towards the reactants, though it is doubtful that this is a significant move because the high temperature should correspond well with stoichiometric mixture contour, and the scattering cross-section of stoichiometric reactants and products differs by less than 0.5%.

The spatial location of the outer and inner reaction fronts are marked clearly by maxima in both OH number density and temperature as seen in Figure 4.9. Asymmetries are due to the imperfections in coflow adaptor symmetry, and thus, asymmetries in the flow. Furthermore, the camera used for Rayleigh scattering was positioned at a different angle than that used for OH PLIF, resulting in different asymmetries observed in the studies. Temperature images were corrected for the non-perpendicular camera placement using an image of a target grid.
Figure 4.9 OH number density (left) and temperature (right) profiles of the solid electrode non-premixed plasma discharge for varying power levels. Inset images depict area of data extraction (25 mm over torch).

The plots confirm the general trends noted in the premixed case; Increases in power lead to increases in OH density and temperature. Again, note that the temperature should be higher than calculated in the fuel rich regions between the maxima due to the use of the cross-section of combustion products. Without microwave power, a flame can be stabilized only if there is no nozzle flow. The increase in OH density is both a function of increasing temperature and flame front movement toward the torch. The temperature is observed to be sufficiently high for combustion at this power; regardless, it can be seen that OH propagation is not very high at even these temperature due to the insufficient mixing of the air and fuel streams.

E. Hollow Electrode and Non-Premixed

Methane was sent through the hollow electrode while air passed through the nozzle around the electrode. The plasma discharge formed on the boundary between the two flows, igniting a non-premixed flame where fuel and air mix. The motivation for a hollow nozzle is mainly due to the
fact that the highest concentration of E-field is at the tip of the electrode and a hollow nozzle
would direct most of the gas directly into the plasma plume. OH number density and temperature
were recorded for different microwave powers and fuel flow rates from 0.2 to 1.0 SLPM (1.4 to
6.9 m/s). Nozzle air flow around the electrode was fixed at 30 SLPM (12.2 m/s). Figure 4.10
contains a graphical representation of the data for a power level of 70 W.

![Graphical representation of data](image)

**Figure 4.10** Images of OH number density [$10^{16}$ cm$^{-3}$] (top), averaged temperature [K] (middle),
and photographs of discharge (bottom), of a non-premixed discharge for varying hollow
electrode fuel flow rates, increasing left to right (70 W).

OH number density images clearly show a flame front along the methane-air interface, as
expected for a non-premixed flame. As fuel flow rate increases, the front stretches, and extends
out of the field of view by 0.6 SLPM. The images show apparent local flame extinction and
greater front curvature at increasing fuel flow rate. Average OH images (not shown) look very similar to the temperature images from the Figure 4.10. Maximum OH radical concentrations range from \((1.5-4.5) \times 10^{16} \text{ cm}^{-3}\) as power is increased from 70 to 130 W but show little dependence on flow rate. For this configuration, the highest median temperatures are not located at the plasma, but downstream, where additional heat from oxidation is released. Increasing the microwave power level has no effect on the median temperature values. As with the solid non-premixed case, there is an under calculation of scattering cross-section, and therefore temperature, of the fuel only center stream and fuel rich mixture near the flame. This would have the greatest effect near the electrode from which the fuel is issued, at maximum doubling the temperature. This would not change the contour of the maximum temperature with much significance but would be expected to decrease the temperature gradient on the inner sides of the flame and increase the center temperature up to a factor of two very near the electrode.

OH number density and temperature profiles at 25 mm above the torch tip are plotted in Figure 4.11. There are two maxima for each line indicating the location of the cylindrical flame front. OH radical concentrations increase significantly at each power level while median temperature remains nearly constant. This indicates that adding microwave energy to this plasma coupled flame can increase OH radical concentration without significant thermal effects, in contrast to observations of solid electrode configurations. Soot formation is observed at low powers, likely due to incomplete combustion. The relatively constant temperature is unique to hollow electrode configuration and contrasts sharply with what was observed in the solid electrode non-premixed case, where the plasma was surrounded by airflow while the fuel flow surrounded the nozzle annularly. These results, along with comparisons of air only plasma and a CH\(_4\)-air plasma coupled flame, suggest gas flow heating is greater in configurations where the plasma is formed in air than at the CH\(_4\)-air interface.
Figure 4.11 OH number density (left) and temperature (right) profiles of the non-premixed hollow electrode plasma discharge for varying power levels. Inset images depict area of data extraction (25 mm over torch, 600 sccm fuel flow rate).

F. Hollow Electrode and Premixed

A second hollow electrode flame was studied by flowing a premixed CH₄-air gas through the electrode. The total premixed flow rate was held constant at 2 SLPM (13.8 m/s) while the microwave power and equivalence ratio varied. Figure 4.12 contains images of OH number density and temperature for a hollow electrode premixed flame at a power level of 100 W for different fuel-rich equivalence ratios. The use of a stoichiometric product mixture scattering cross-section causes an under calculation error in temperature up to 9%, 23%, 33%, and 40% for equivalence ratios φ = 2, 4, 6, and 8, respectively. This maximum error due to incorrect composition would occur above and very near the hollow electrode, as it requires that no surrounding air has mixed or diffused into the reactants. The photographs show the flames have a long conical structure that lengthens as the equivalence ratio increases. The OH profiles show a cylindrical flame front, thicker than that observed in the non-premixed case, with a maximum concentration range of (1.0-4.0) ×10¹⁶ cm⁻³. OH number density is inversely related to
equivalence ratio, with maximum concentrations 10 to 20% greater for flames with an equivalence ratio of 2.0 than 8.0. Median temperature images also show a decrease in temperature as fuel fraction increases, ultimately approaching temperatures seen in the non-premixed flame as the fuel fraction becomes very large. The hollow electrode with premixed flow allows for reliable ignition and stable combustion of a very rich flow without sooting at velocities greater than 10 m/s.

Figure 4.12 Images of OH number density \(10^{16}\) cm\(^{-3}\) (top), averaged temperature [K] (middle), and photographs of discharge (bottom), of a premixed discharge for varying hollow electrode flow equivalence ratio (100 W).
Figure 4.13 contains plots of OH concentration and temperature 25 mm above the torch tip. Peak OH number density scales nearly proportional to microwave power. A 50% increase in power, from 80 W to 120 W, yields a 60% increase in peak OH number, whereas temperature increases by about 15%. Also plotted are temperature profiles for 3 downstream locations for a premixed flame with an equivalence ratio of 6.0 and 90 W of microwave power. It is observed that the median temperature increases moving away from the torch. The plasma ignites the flow, but most of the heat release occurs downstream as the fuel continues to oxidize. Recall that the temperature in the middle of the curve is under calculated due to composition assumption. This has the greatest effect on the profile for 6.5 mm above the torch, and the corrected temperature could be (at most) $1.33 \times$ the plotted value in the center region.
Figure 4.13 OH number density (left) and temperature (center) profiles of the premixed hollow electrode plasma discharge for varying power levels with inset images depicting area of data extraction (25 mm over torch). Temperature profile at three downstream locations in a flame with an equivalence ratio of 6.0 and microwave power level of 90 W (right).
Temperature profiles suggest plasma induced initial fuel cracking and continued oxidation downstream. This configuration offers the greatest advantage of targeted energy deposition into a fuel rich flow to initiate combustion through CH$_4$ decomposition. Studies of the kinetic mechanisms of plasma-based CH$_4$ conversion have produced data outlining in-situ fuel reformation pathways of interest for plasma assisted combustion [112, 113]. A non-equilibrium plasma facilitates additional pathways for CH$_4$ decomposition, including CH$_4$ excitation via electron impact and subsequent breakup. Energetic nitrogen species actively initiate oxidation: CH$_4$+N$_2$(A, v =1) or CH$_4$+N$_2$(X, v >20)→CH$_3$ +H +N$_2$.

G. Power into Heating

Analysis of gas temperature near the plasma discharge was conducted to investigate the thermal nature of the discharge. The heating of the gas flow can be calculated from the increase in temperature as power increases and expressed as a heating power using the equation

$$ P = \dot{V}c_p \rho \Delta T $$

(4.8)

where $\dot{V}$ is the volumetric flow rate, $c_p$ is the specific heats, $\rho$ is the density, and $\Delta T$ is the change in mean discharge temperature weighted for the three dimensional geometry. The gas heating can be expressed as a percentage of an increase in microwave power, where $\Delta T$ is evaluated between the two power levels.


Table 4.1  Plasma heating in an air discharge and plasma assisted combustion, calculated using the change in temperature over a power range interval. The total flow rate is 5 LPM.

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<td></td>
<td>Air</td>
<td>CH4/Air</td>
<td>Air</td>
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<td>60 to 70</td>
<td>134.24</td>
<td>83.05</td>
<td>6.92</td>
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<tr>
<td>70 to 90</td>
<td>258.71</td>
<td>180.35</td>
<td>10.52</td>
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<tr>
<td>90 to 110</td>
<td>216.26</td>
<td>86.48</td>
<td>7.09</td>
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<td>110 to 130</td>
<td>170.57</td>
<td>59.72</td>
<td>4.83</td>
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Table 4.1 contains the increase in temperature, heating power, and heating as a percent of the increase in microwave power for both an air-only discharge and plasma assisted combustion at equivalence ratio φ =1. The percent power into heating is much higher in the air-only discharge than in the PAC discharge, where a very large portion of plasma energy couples as non-thermal effects. As power is increased, the expression of plasma energy as heat decreases. From this data, it is demonstrated that the thermal effects are lesser in a plasma enhanced flame than in air, and lesser at greater microwave powers. The power into heating the air plasma is about four times greater than for PAC over the same power interval. At these conditions, the microwave energy per molecule is highest, and non-thermal effects, including fuel breakdown through electron impact dissociation and excited state species interaction, are maximized in relation to heating interactions, such as relaxation and energy transfer. This introduces the need to assess the value in using greater plasma energy to increase non-thermal effects.
4.1.5 Conclusion

A microwave waveguide plasma source has been developed to enable laser and optical diagnostics of direct coupled plasma enhanced flames. OH PLIF and planar Rayleigh scattering thermometry are used to examine four distinct premixed and non-premixed CH$_4$-air plasma enhanced flames. The plasma discharge and flames are produced at the torch nozzle exit, allowing for the optical accessibility for planar diagnostics. The different nozzle configurations and flame geometries are compared and considered for energetically enhanced combustion applications.

The highest temperatures are observed in the solid electrode non-premixed flame, where the plasma is located within the center airstream, while significantly lower temperatures are measured in the premixed flame with equivalent total flow rates. The non-premixed flame would be best suited for applications requiring rapid gas flow heating and high gas temperatures (greater than 3000 K). The cooler premixed configuration is much more efficient in terms of combustion power to MW plasma power, as high flowrates of CH$_4$-air are well stabilized by the plasma discharge. In the hollow electrode configurations, temperature is much lower and peaks well downstream of the plasma. The hollow nozzle non-premixed case produces sooting flames at low fuel flow rates, and increasing flame front curvature with signs of frequent local extinction as the fuel flow rate increases. Using a rich premixed flow through the hollow electrode maintains low plasma gas heating, but eliminates sooting and greatly reduces flame front discontinuities. We believe this is the most promising configuration for non-thermal enhancement of high velocity CH$_4$-air flames, as microwave energy can be used more efficiently via CH$_4$ decomposition through mechanisms facilitated by plasma-flame chemistry. The application requirements will ultimately dictate the optimal nozzle configuration.
CHAPTER 5  OH PLIF

5.1 10-kHz OH PLIF in a Supersonic Wind Tunnel Cavity Flameholder

The results of an experimental investigation of a supersonic cavity combustor using high-frequency planar imaging diagnostics are presented in this work. Specifically, 10-kHz OH planar laser-induced fluorescence (PLIF) is used to capture flame geometry and dynamics within a cavity flameholder in a supersonic crossflow (Mach 2). The combustor behavior is examined for direct fuel injection into the cavity over a range of fuel flow rates. The 10-kHz PLIF used here is the first integration of sustained high-frequency PLIF imaging in a supersonic combustor, and while the frame rate is not sufficiently fast to resolve flame dynamics in general, these measurements allow the acquisition of a large and statistically significant database and represent a first step towards time resolution within a high-speed flowfield.

5.1.1 Background

Supersonic combustion requires the use of a specialized combustor geometry to maintain stable operation, due to short aerodynamic time scales relative to reaction time scales. Wall cavity combustors have been demonstrated as a viable option for flameholding and ignition in a dual-mode scramjet engine [92, 114]. Studies have investigated the practical application of various cavity geometries for scramjet engine flameholding, examining i) fuel-mixing and rich/lean blow-off [115, 116]; ii) recirculation zones, velocity fields, and shear-layer interaction [94, 117]; iii) pressure oscillations, cavity drag, and shock propagation [114]; and iv) numerical simulation [118]. Different fuel injection strategies have also been explored, resulting in an understanding of the importance of fuel injector placement [90, 93, 115, 119, 120].
5.1.2 Experimental Setup

The experiments presented here were performed in the direct-connect supersonic research facility at Wright-Patterson Air Force Base (Research Cell 19) [85]. Continuous air flow is conditioned by a settling chamber, accelerated through an asymmetric nozzle to Mach 2, and then passed through a constant-area isolator section measuring 50.8-mm high and 152.4-mm wide before entering the test section where the floor diverges at 2.5°. The recessed wall cavity is 76.2 mm downstream from the start of the test section and measures 16.5-mm deep and 66-mm long (to the midpoint of the closeout ramp), while spanning the tunnel width of 152.4 mm. The upstream rearward-facing step is a 90° corner while the closeout ramp is at 22.5° relative to the duct floor. Ethylene (C₂H₄, chemically pure grade) is injected through a spanwise row of 11 fuel injectors in the ramp, such that the fuel jets are parallel to the floor and opposing the freestream flow direction, following the recirculation pattern within the cavity. Additional fuel can be injected into the main tunnel flow upstream of the cavity through a 25-mm wide floor slot in the spanwise center of the floor. Fused-silica windows allow for optical access from both sides and the top of the test section. Additional details and illustrations of the cavity geometry can be found in Ref. [94] by Tuttle et al. and Ref. [90] by Hsu et al. For this study, the respective total temperature and pressure were 611 K and 483 kPa.
Figure 5.1 illustrates the setup of laser and optical equipment. OH radicals were excited using the $Q_1(7)$ transition of the $A^3\Sigma^+-X^2\Pi (1,0)$ band. A diode-pumped solid-state Nd:YAG pulsed laser (EdgeWave Innoslab IS12II-E) fired at 10 kHz, produces 532-nm radiation using an LBO frequency doubling crystal. A tunable dye laser (Sirah CREDO), designed for high-frequency, high-power operation, produced output radiation at 566 nm using rhodamine 590 dye (Exciton). An integrated frequency control unit houses a BBO crystal for frequency doubling to 283 nm; for this experiment output pulse energy was about 120 µJ. Lenses expand the beam streamwise into a 70 mm wide sheet and produce a narrow waist at the probing volume. The laser sheet passes through the center of the top tunnel window such that imaging is along a symmetric plane. A fused-silica window placed in the beam path is used to reflect a small portion of the UV radiation
over a premixed test burner, allowing for excitation scans (and wavelength verification) using a monochromator fitted with collection optics and a photomultiplier tube. A high-framing-rate camera (Photron SA-5) and image intensifier (LaVision HS-IRO) equipped with a 45-mm, f/1.8 UV lens (Cerco) and a high-transmission (>80% at 310 nm) bandpass filter (Custom fabrication – Laser Components GmbH) records the OH fluorescence in the cavity combustor from transitions of the \( A^2\Sigma^+ - X^2\Pi \) (1,1) and (0,0) bands; the intensifier gate time was set to 200 ns, and this was sufficient to block the background OH chemiluminescence. Image sizes were 1024x680 pixels, with a field of view of about 71 (long) \( \times \) 44 (high) mm at a resolution of 69x69 µm per pixel. Signal digitization was at 12 bits, equivalent to 4095 grey levels. The camera was equipped with 8 GB of internal memory, which allowed measurement times as long as 822 ms. Thus, about 5400 images could be collected for each combustor run, whereas a 10-Hz Nd:YAG-dye laser system provides only 200 images for a typical combustor run. A pulse generator (Berkeley Nucleonics Corporation) triggered the laser and camera systems at 10 kHz, while a camera output triggered the image intensifier gating. Acquisition was controlled through the camera software application Photron Fastcam Viewer 3 on a control PC. Collected data for each acquisition was stored in the camera RAM, allowing for playback and review before upload to the control PC via gigabit Ethernet.

5.1.3 Results and Discussion

The high repetition rate laser and imaging systems produced data that exhibit excellent signal-to-noise ratios, high spatial resolution, and the extension of cavity PLIF studies into the multi-kHz regime. Sequential image sets for four fueling conditions are provided in Figure 5.2, along with corresponding ensemble average images. A low-power flame is sustained at an ethylene flow rate of 41.2 SLPM (standard conditions corresponding to 273.15 K and 1 standard atm),
spanning from the fuel injectors to as far as the upstream cavity step. A high degree of flamefront (i.e., OH surface) wrinkling and curvature is observed, as well as in-plane rotation consistent with the primary recirculation expected for this geometry. Combustion appears to occur in distinct periodic events every 0.5 to 1.2 ms, with OH appearing near the fuel injectors then moving through the cavity as a wrinkled and often discontinuous region. The flame area is greatest 30 to 40 mm from the upstream step and decreases near the step. As the fuel flow rate increases, the spatial variation in the OH decreases in the primary flame zone, as identified by the averaged images in Figure 5.2. Increasing the fuel flow to 63.7 SLPM increases the combustion volume near the ramp, shifting the primary flame zone slightly downstream and results in a more robust flame near/in the cavity shear layer. The additional heat release and expansion of cavity gases shifts the shear layer upwards. It should be noted that due to rapid fuel-air mixing rates within the cavity, soot precursors are not formed (soot emission is not observed) in sufficient quantities to generate LIF competitive with that from OH [90, 93, 116, 119].
Figure 5.2  Average and sequential images for a) medium (41.2 SLPM), b) medium-high (63.7 SLPM), c) high (112.6 SLPM), and d) very-high (162.8 SLPM) fuel flow rates.
Figure 5.3 Average and sequential images for a) low fuel (31.6 SLPM), and b) slot fuel (50.8 SLPM + 85.8 SLPM). The low fuel flow rate corresponds to the lean blow-off condition.

Further increases in fuel injection reduce the intensity of the shear layer flame and continue to shift the primary flame zone downstream toward the cavity close-out ramp, until a flow rate of 162.8 SLPM. At this condition, the flame density is highest, and the flame and products occupy most of the near-ramp region. Signal level decreases at higher flow rates while the profile changes very little. Combustion was sustained up to 214.4 SLPM, though a rich blow-off limit was not established. At 10 kHz in-plane flame convection and propagation may be expected to be well characterized by OH PLIF, due to the relatively low cavity velocities [94], but out-of-plane motion limits the tracking of features from frame to frame. Of course, turbulent flows have strong three-dimensional effects, and any planar technique will be limited in its ability to resolve fluid motion because of this. Still, qualitative observations of highly transient flame behavior can be made. Flame (OH) convection becomes more difficult to identify and follow from frame to frame as upstream OH seems to appear randomly and movement through the cavity is unclear, likely due to increasing out-of-plane motion. Combustion in the upstream half of the cavity is
infrequent and discontinuous from the bulk flame. At high fueling rates, the OH signal indicates stable combustion from frame to frame in the downstream half of the shear layer and near the fuel injectors. The general trend is marked unsteadiness and flame discontinuities at low combustion powers and greater flame area stability and continuity in the imaged plane as the fuel rate is increased until maximized at 162.8 SLPM.

Sequential image sets and ensemble average images for lean blow-off and slot fueling are provided in Figure 5.3. Lean blow-off (LBO) occurred at 31.6 SLPM, as intermittent regions of combustion are seen between the ramp and upstream step. At LBO (31.6 SLPM), similar periodic flame convection occurs, though the high signal region is now absent. Additional ethylene fuel was injected into the main tunnel flow upstream of the cavity, through a low-angled slot located about 180 mm upstream of the cavity step, while the cavity fuel flow rate was maintained at 50.8 SLPM. For $Q_{\text{slot}} = 85.8$ SLPM, the additional fuel through the upstream slot decreases the primary flame area, most prominently in the shear layer flame.

Consideration of previous work examining this cavity provides additional insight into the complex flow structure. Tuttle et al. observed that combustion produced broadening of the shear layer and attenuated unsteadiness of both the velocity and vorticity in the cavity, shear layer, and at the stagnation of the shear layer against the ramp. Averaged vorticity profiles showed greater variance as fuel flow was increased and suggested an intermittently stable “lifted non-premixed flame” along the shear layer with a very strong out-of-plane secondary flow. While we cannot verify the vorticity and variance findings, our data does depict a seemingly lifted flame, shifted back along the streamline, for similar fuel flow conditions (63.7 and 96.2 SLPM).
5.1.4 Conclusions

This study presents OH imaging over a wide range of fuel flow rates for a directly fueled cavity at a high sampling rate wherein highly transient complex flame dynamics were captured. This effort serves to better describe cavity flameholding and verify mechanisms suggested by diagnostics at lower interrogation rates. It is now possible to very quickly record statistically significant numbers of sequential images to better characterize the mean and variance of OH distribution within a turbulent flame. Data acquisition yielding thousands of high resolution images can be performed in less than 1 second, and thus the study of many combustion cases is now possible. Furthermore, investigation of time dependent phenomena, such as the effect of transient heating of flameholder surfaces and flame-acoustic coupling, is also possible.

In the present work, location of a stable flame, inferred from OH PLIF images, shifts based on fuel flow rate (with cavity fueling) even with all other parameters held constant. At low fuel flow rates, flame position is determined by the primary recirculation zone, with bias towards the ramp and the shear layer. Flame convection from the main combustion region towards the upstream step occurs in periodic events, repeating approximately every 1 ms. With increasing fuel flow, the shear layer thickens, and a shear layer flame is clearly established, while the bulk of the combustion shifts downstream. Distinct flame convection from frame to frame becomes ambiguous due, presumably, to increasing out-of-plane effects. Higher fueling rates weaken the shear layer combustion and move the flame location towards the ramp, until the region of combustion is nearly attached to the injectors. Finally, it is noted that advances in diode-pumped laser and CMOS camera technologies will broaden the range of phenomena that can be studied, including ignition, blowout, etc.
5.2 10-kHz OH PLIF and OH* Chemiluminescence of Microwave Plasma Torch

This study examines the structure of microwave enhanced flames through 10-kHz imaging. High repetition rate laser diagnostic methods are implemented to simultaneously record two-dimensional images of OH laser-induced fluorescence and chemiluminescence within an atmospheric plasma-enhanced flame. Collecting both OH planar laser-induced fluorescence and chemiluminescence allows for observation of OH radicals in the plane of the thin laser sheet as well as volume-integrated excited state emission. A tunable, microwave waveguide plasma source—operating at 2.45 GHz and delivering 90 to 130 W to the flowfield—ignites and sustains a CH4/air flame, while laser-induced fluorescence and chemiluminescence are acquired at a sustained framing rate of 10 kHz, utilizing two intensified CMOS cameras and a synchronized laser. Multiple geometries and flames (premixed and nonpremixed) are studied by adjusting gas flow compositions and the plasma applicator nozzle components. A stoichiometric premixed flame configuration produces a divergent flame with large scale fluctuations and vortex shedding into ambient air and is capable of feedstock flow velocities greater than 20 m/s for combustion-to-plasma power ratios greater than 10:1. Another arrangement produces plasma along the initial mixing layer of a non-premixed flame, yielding a thin cylindrical reaction zone of coincident chemiluminescence and fluorescence. Replacing the fuel with rich premixed gases produces a narrow conical flame anchored by the circular plasma discharge with little flamefront fluctuation. The high-speed diagnostics capture OH signals in cinematic sequences, providing new understanding of the plasma-assisted flame holding mechanism and allowing for tracking of individual flow feature development.
5.2.1 Introduction

Modern combustion engineering is often faced with the task of extending the limits of the current state-of-the-art. Chemical energy conversion is to be efficient, harnessing more energy while burning less fuel, sometimes even a new, poorly characterized fuel. Pollutants and otherwise undesirable byproducts must be monitored and a reaction environment maintained so as to limit the offending chemical pathways. Energetically enhanced combustion introduces new kinetic mechanisms while altering existing reaction rates and sensitivities. Applications rarely feature the well-defined laminar flames of laboratory test burners that facilitate high-quality diagnostics and modeling. Instead, the challenge is to produce reliable combustors for turbulent, high velocity flows that are prone to extinction and are highly transient in nature. It is in this effort that the progressing capabilities of laser diagnostics open new possibilities of understanding the chemistry and fluid dynamics of complex reacting flows.

The recent development of high-speed tunable dye lasers and intensifiers creates the opportunity for selective tuning and sustained operation like that of the conventional 10 Hz systems. Such a system, complemented by additional lasers and cameras for simultaneous stereoscopic PIV, provides the tools for the study of transient phenomena in turbulent combustion through time-resolved flamefront tracking and three-component flowfield construction. These techniques have been demonstrated for jet diffusion flames [61, 62, 64-66], opposed jet flames [51, 67], and swirl-stabilized gas turbine combustion [64, 68-70] at framing rates of 1.5 to 10 kHz, capturing the great significance of quasi-four dimensional flowfield measurements on flame stabilization investigation.

The present study utilizes a frequency-doubled, diode-pumped, 10-kHz Nd:YAG laser to pump a frequency-doubled, high-speed tunable dye laser to excite OH radicals in a plasma-enhanced
Flame. Fluorescence and chemiluminescence from a microwave-plasma-ignited and stabilized flame are simultaneously recorded by intensified CMOS detectors at 10,000 frames per second (fps) to yield new understanding of the plasma-assisted flameholding mechanism and allow tracking of individual flow features.

5.2.2 Experimental Setup

A. Microwave Plasma Source

Plasma enhanced flames were produced using a tunable microwave waveguide and a coaxial, plasma-applicating torch nozzle designed in collaboration with 'Amarante Technologies' of Santa Clara, California. The unique nozzle facilitates plasma formation at atmospheric pressures, external to the apparatus and thus optically accessible, and centered over the tip of the flameholding nozzle. Initial studies examined flame structure and behavior through 10-Hz OH PLIF and Rayleigh scattering thermometry, as power input, flow rates, and equivalence ratio were varied [121-123]. Further experimentation explored the OH radical and temperature profiles for various nozzle configurations that produce nonpremixed and premixed flames with differing plasma coupling modes. Here, we will examine the flowfield using high-speed imaging of OH PLIF and chemiluminescence, simultaneously recorded at 10 kHz; while it was possible to record several thousand frames at a time, limited only by camera onboard memory, typically only 1000 images were captured and saved.

The microwave plasma source (MPS) has been introduce in Chapter 4 and diagramed in section Figure 5.4. A continuous power generator (Alter Power Systems) regulates a magnetron head (National Electronics) emitting into the WR284 waveguide at an output of 360 W to 2 kW at 2.45 GHz in the TE10 mode. Incident power is measured near the magnetron using a directional
coupler. An isolator, consisting of a circulator and water-cooled dummy load with integrated coupler, facilitates absorption and measurement of reflected power. The waveguide is tuned using a three-stub tuner (National Electronics) and sliding short (Gerling), so that the reflected power can be adjusted. The plasma applicator nozzle (Amarante Technologies) allows for plasma and flame coupling, functioning as a coaxial waveguide and torch. The magnetron was operated at 360 W for all cases presented. As this is more energy than needed for ignition and stabilization, the waveguide is detuned to reflect a portion of the incident power. The plasma power reported in the results is the difference between the fixed incident power (360 W) and the reflected power as measured at the dummy load. This absorbed microwave energy value is also used to calculate the ratio of net deposited power to combustion power.

Figure 5.4 Microwave plasma source diagram (top left) and plasma source photograph (right). The torch can be configured with a solid or hollow electrode and has been modeled using the COMSOL Multiphysics RF Module (bottom left, left to right). The false color describes the E-field distribution, greatest at the electrode tip.
The plasma applicator nozzle can be configured to produce premixed and non-premixed flames, while using a solid electrode or a hollow grounded electrode (shown in Figure 5.4). Three configurations are presented in this study: 1) a solid electrode with premixed flame, 2) a hollow electrode with non-premixed flame, and 3) a hollow electrode with premixed flame. As illustrated in Figure 5.4, the hollow electrode provides a second gas flow path for fuel or premixed flow while air flows outside the electrode. A Teflon insert holds the solid electrode in position while preventing the nozzle from entering the waveguide. Furthermore, to prevent arcing, a short quartz tube is inserted flush with the nozzle tip. The multiple nozzle configurations are compared in terms of flameholding, flame geometry, and temperature to study the effectiveness of MW plasma to assist combustion.

**B. Laser Diagnostics**

OH radicals were excited using the $Q_1(8)$ transition of the $A^2\Sigma^+ - X^2\Pi (1,0)$ band [3]. An Nd:YAG laser (EdgeWave Innoslab), frequency-doubled to 532 nm, pumped a dye laser (Sirah CREDO) at a rate of 10 kHz. The integrated frequency conversion unit doubled the dye laser beam from 566 nm to 283 nm. The laser beam was expanded into a sheet using cylindrical concave and spherical convex UV lenses. Power was measured at 4.8 W, corresponding to 480 μJ per pulse. Fluorescence was collected at 90 degrees using a high-speed camera (Photron SA-5) and intensifier (LaVision HS-IRO) equipped with a 100 mm, f/2.8 UV lens (Cerco) and filter. OH chemiluminescence was recorded with a second intensified camera (Photron SA-5 and Hadland Imaging intensifier) opposite the PLIF camera, as shown in Figure 5.5; the chemiluminescence camera was fitted with a 45 mm, f/1.8 UV lens (Cerco) equipped with a bandpass filter for OH emission. To achieve 10 kHz framing rates, the array size was set to 768 × 768 pixels.
Laser, cameras, and intensifiers were synchronized using external pulse generators. The PLIF intensifier was gated around the laser pulse ($\Delta t_{gate} = 200\,\text{ns}$) to capture fluorescence, while the chemiluminescence intensifier gate ($\Delta t_{gate} = 10\,\mu\text{s}$) was delayed slightly to avoid capturing fluorescence but maintain approximate simultaneity with respect to the PLIF measurement. Estimates of OH number density are derived from previous measurements in this burner; in that work, a Hencken burner, calibrated through absorption, was used to correlate signal intensity with number density. Temperature fields are also referenced from the same study [122, 123]. Chemiluminescence is reported only in terms of arbitrary units.
5.2.3 Results and Discussion

All data presented are from CH$_4$-air plasma-enhanced flames. The ignition procedure was to flow the fuel and air, then ignite the flame by increasing effective microwave power to the nozzle tip, through adjustment of the three stub tuner, until breakdown and plasma formation. In all cases the flame is not stable without the plasma discharge; extinguishing the plasma discharge results in flame blow-off. Flow rates are measured in SLPM (standard liters per minute), following the IUPAC standard temperature and pressure as 273.15 K and 1 atm. The microwave (MW) power is defined as the difference between measured incident and reflected power. OH PLIF quantification followed the same methods as for 10 Hz studies (see section 4.1 Microwave Plasma Torch 10 Hz diagnostics).

A. Premixed Flame and Solid Electrode

A plasma plume is produced on the end of a solid tungsten electrode held in place by a Teflon spacer. Premixed flow enters the nozzle from the side and exits through the tip, isolated from the waveguide by the Teflon spacer. The flow ignites and a flame stabilizes quickly into a jet structure, by eye appearing blue with blurred and poorly defined edges. The plasma is visible as a violet cone, 3 to 5 mm in height and extending slightly in diameter past the electrode edges.

Sample data are presented in Figure 5.6, with PLIF and chemiluminescence image sequences for MW power settings of 90 and 110 W. The flow is stoichiometric, with a total flow rate of 15 SLPM, yielding an average exit velocity of 16.5 m/s and an absorbed MW power to combustion power ratio of 0.11 to 0.13. The PLIF images at 90 W reveal detachment of the flame base from the torch; the signal is very weak just above the plasma and increases sharply 10 mm from the nozzle tip. The strongest signal in the PLIF images is from the plasma plume, which emits many UV wavelengths, including OH emission within the filter's high transmittance band. This is not
evidence of combustion, as the images demonstrate the flamebase to be lifted, but only strong plasma emission. Regions of expanding low signal levels within the flame boundaries are apparent in both PLIF and chemiluminescence images, a verification of interior flamefronts burning outwardly. Other pockets are fresh reactants, shrinking as the enveloping flame propagates inward. Temperature fields obtained in our previous work with Rayleigh scattering, where temperatures were derived by median filtering pixel values for the ensemble of images, depict similar flame structure (see section 4.1 Microwave Plasma Torch 10 Hz diagnostics) [122, 123]. Temperatures reach 2500 K in the plasma plume and 2200 K in the flame. An increase in MW power, from 90 W to 110 W, results in a larger and more uniform flame volume and an increase in plasma plume length. The distance between plasma emission and flame OH-LIF is reduced, but the flame base appears to maintain a height of approximately 10 mm from the torch. Regions of low PLIF and chemiluminescence signal level are smaller and less frequent, vanishing more quickly (i.e. upstream) compared to those observed in the 90-W discharge, and the plasma temperature increases by 50 to 100 K. Experiments to produce a nonpremixed flame, with fuel flowing around the outside of the nozzle and air through the nozzle, resulted in high temperatures in the air-plasma region and poor combustion performance.

B. Nonpremixed Flame and Hollow Electrode

Methane was sent through the hollow electrode while air passed through the nozzle around the electrode. The plasma discharge formed on the boundary between the two flows, igniting a nonpremixed flame where fuel and air mix. The motivation for using a hollow nozzle is primarily that the highest E-field is around the top of the electrode (as illustrated in Figure 5.1), such that a hollow nozzle would produce a plasma discharge at the boundary layer of an interior and exterior flow. Figure 5.7 contains image sequences of OH PLIF and chemiluminescence for MW power levels of 90 and 110 W, with a fuel flow rate of 0.6 SLPM (4.2 m/s) and an absorbed
MW power to combustion power ratio of 0.25 to 0.31. Nozzle air flow around the electrode was fixed at 30 SLPM (12.2 m/s).

Figure 5.6 OH PLIF and chemiluminescence image sequences for a premixed plasma coupled flame using the solid electrode.
Figure 5.7 OH PLIF and chemiluminescence image sequences for a nonpremixed plasma coupled flame using the hollow electrode.

The OH PLIF images clearly illustrate a flamefront along the fuel-air boundary. The flame is initiated around the cylindrical tip of the hollow electrode, near the mixing layer, anchored by the plasma discharge. Chemiluminescence shows regions of OH* with the same outer profile,
demonstrating a high degree of temporal correlation between data. The OH* signal is greatest along the flamefront 15 to 20 mm above the torch where the flamefront curvature increases. Temperature fields revealed the highest average temperatures are not located in the plasma, but downstream, where additional heat from oxidation is released. Increasing the microwave power level from 90 to 110 W has no measureable effect on the temperatures. There is an under calculation of scattering cross-section, and therefore temperature, of the fuel only center stream and fuel rich mixture near the flame. This would have the greatest effect near the electrode from which the fuel is issued, at maximum doubling the temperature. This would not change the contour of the maximum temperature with much significance but would be expected to decrease the temperature gradient on the inner sides of the flame and increase the center temperature up to a factor of two very near the electrode.

The 10-kHz diagnostics reveal transient characteristics of this plasma-coupled flame. What appears to be a continuous circular plasma discharge is actual several discrete plasma plumes moving rapidly around the electrode tip. This can produce momentary liftoff of part of the flame, as observed in the 90-W OH PLIF images. Further downstream, local extinction is apparent, where there is a break in the flame front followed flame propagation inwards, collapsing the flame and producing a distinct flame. These extinctions are most likely caused by mixing layer vorticity propagation, observable in extending sequences as oscillations in the radial flamefront position, increasing in amplitude as the move downstream. Without the high viscosity reaction from between the fuel and air flows, the front propagates quickly into a now partially premixed region, encapsulating a fuel rich region. Of course, in the absence of any flowfield and out of plane OH PLIF data, analysis of these events is limited to speculation. Comparing the two power levels, the flame with 110 W of MW input power does not exhibit the partial liftoff observed in the 90-W flame, evident in the short sequences in the figure.
C. Premixed Flame and Hollow Electrode

A premixed flame was produced by flowing air and fuel through the hollow electrode. The plasma discharge formed as ring around the tip of the electrode, the same structure observed for the non-premixed configuration. The mixture was held to a total flow rate of 2 SLPM (corresponding to a jet exit velocity of 13.8 m/s) and a very rich composition at an equivalence ratio of 6.0. Nozzle air flow around the electrode was again fixed at 30 SLPM (12.2 m/s) yielding an absorbed MW power to combustion power ratio of 0.15 to 0.16. Figure 5.8 contains image sequences of PLIF and chemiluminescence for 100 and 110 W of MW power.

The flame is shaped as a narrow cone, similar to a premixed Bunsen flame, but stretched axially by the high gas velocity. The plasma discharge anchors the flame to the electrode as the flamefront extends nearly parallel to the flow before converging to a tip approximately 50 mm above the torch. OH PLIF reveals a flamefront extending from the discharge along the mixing layer between the air stream and rich premixed gas. There is no clear evidence of significant ground-state OH radicals in the reactant region inside the flame. OH chemiluminescence captures the same flame structure.

The hollow electrode with premixed flow allows for reliable ignition and stable, non-sooting combustion of a very rich mixture at velocities greater than 10 m/s. The flame length increases with greater fuel fractions and decreases with greater MW power. Flame temperature peaks 25 mm above the torch at approximately 1600 K. Increasing the MW power produces more uniform OH PLIF and chemiluminescence distributions, but has little effect on the median temperature field. Near the plasma discharge, average temperatures are well under 1000 K. This suggests plasma-induced fuel cracking and initiation reactions facilitated by the plasma-flame coupling and continued oxidation downstream of the plasma. However, the scattering cross-section of stoichiometric and complete combustion products was applied. This works well enough for
Figure 5.8 OH PLIF and chemiluminescence image sequences for a premixed plasma coupled flame using the hollow electrode.

Premixed flames near $\phi = 1$ as there is very little change in the cross-section. In the case presented here, the flow is very rich ($\phi = 6$, it might be called partially premixed) and so the use of a stoichiometric product mixture scattering cross-section causes an under calculation of
temperature by as much as 32.7%. This maximum error due to incorrect composition would occur above and very near the hollow electrode, as it requires that no surround air has mixed or diffused into the reactants. Taking this into account, a corrected is the average temperature near the plasma discharge is certainly under 1333 K. Figure 5.9 shows the temperature and excited OH development along the flame. Studies of the kinetic mechanisms of plasma-based CH₄ conversion have produced data outlining in-situ fuel reformation pathways of interest for plasma assisted combustion [112, 113]. A non-equilibrium plasma facilitates additional pathways for CH₄ decomposition into CH₃ and H, including CH₄ excitation via electron impact and subsequent breakup or reaction with an energetic body. OH radicals are quickly formed through chain-branching reactions containing O₂ and H. The hollow electrode premixed torch produces a very stable flame at high strain rates without the excessive flow heating evident with the other configurations.

**Figure 5.9** OH chemiluminescence (left) and temperature profiles (right) of the hollow electrode premixed flame at different heights above the torch.
5.2.4 Conclusion

The advent of high-speed diagnostic equipment, including kHz-rate CMOS cameras, image intensifiers and tunable lasers, has enabled signal collection at framing rates sufficient to observe transient features of reacting flows. This study has demonstrated simultaneous OH PLIF and chemiluminescence at 10 kHz applied to an energetically enhanced flame. Cinematic sequences show smooth flow development over the 100 ms recording period (1000 frames). This allows for an increased understanding of the flame dynamics and a better performance comparison between the different nozzle flow and electrode options available for the tunable, waveguide-based, microwave plasma source. Magnetron output power was fixed while tuning components were used to adjust the effective absorbed power. Flames were ignited and stabilized by the plasma discharge at velocities greater than the natural blow-off limit. The plasma-coupled nonpremixed flame using a hollow electrode showed signs of brief partial liftoff and flame extinction. Employing a fuel-rich mixture produces an exceptionally stable flame, as is evidenced by the stability of the OH PLIF field. With this configuration, temperature and OH* emission increase downstream from the torch exit. It is proposed that the discharge initiates combustion through fuel breakdown and the production of active radicals, ions, and high-temperature free electrons, while the most exothermic reactions occur downstream. In applications where high temperature is desirable, the solid electrode (with premixed gases) is preferable. This configuration allows for higher power flames than other configurations (into the kW range) at mean flow velocities of over 20 m/s and average temperatures near 2500 K. Gas flow heating is greater for non-premixed flows with air-only plasma feedstock, producing hot regions up to 3500 K; thus, this configuration is well suited for uses necessitating rapid heat generation, ignition, and flame development.
5.3 Demonstration of 50-kHz OH PLIF

The purpose of this study is to demonstrate the first use of OH PLIF at a rate of 50 kHz using a continuously operating laser system, where the only limitation for data record length is the camera memory size. An Nd:YAG-pumped dye laser system is used for OH excitation and an intensified CMOS camera for fluorescence imaging. Results are presented for a stable Hencken burner flame and a turbulent flame produced by a transient-arc DC plasmatron.

5.3.1 Experimental Setup

A. Laser System

Figure 5.10 diagrams the experimental setup. A diode-pumped, Q-switched Nd:YAG laser (EdgeWave Innoslab HD40II-ET) operating at 50 kHz pulse repetition rate is frequency doubled to produce up to 200 W at 532 nm. The 532-nm beam with pulse width of 8 ns pumps a tunable dye laser (Sirah CREDO), modified for the high power and repetition rate of the Nd:YAG laser. A portion (~8%) of the pump power is reflected by a beam splitter to the grazing-incidence resonator assembly of the CREDO. Here, tunable laser pulses with a spectral bandwidth of 0.06 cm⁻¹ and an average power of up to 4 W at a wavelength of 566 nm are generated. An optical telescope with different magnification factors in the tangential and sagittal planes adapts the spatial beam profile for optimum amplification of the laser pulses in a separate dye cell. The dye-cell assembly itself is made of stainless steel for safety reason. Two cylindrical lenses form the residual pump laser radiation to a focused line on the amplifier cell. The pulse duration of the generated laser pulses is ~6 ns, and the beam diameter ~1.5 mm. Rhodamine 6G dye (Exciton) is used within both the oscillator and amplifier dye cells. Dye concentration in the amplifier is lower than with the CREDO designed for 10-kHz pumping. A variable attenuator assembly, consisting of a half-wave plate with servo controlled rotational mount and a high-damage-
threshold polarizing beam splitting cube, is placed between the Nd:YAG and dye lasers, allowing for control of power into the dye laser while the Nd:YAG laser remains at a constant output.

Figure 5.10  Laser and imaging setup

The dye beam is frequency doubled to 283 nm within the dye laser using β barium borate (BBO) crystals and isolated using a wavelength separation unit (an assembly of four Pellin Broca prisms.) To minimize temperature gradient effects, which disturb the phase-matching along the beam path inside the nonlinear crystal, a dual-crystal setup is employed; additionally, both
crystals are temperature controlled, with heaters integrated into the crystal mounts. With the help of this technique, the CREDO dye laser system generates stable average powers of more than 7W at 283 nm at a repetition rate of 50 kHz. The mounts for the two crystals are mechanically linked so that both rotate together, but in opposite directions, such that both doubling and compensation is achieved. The system is tuned by first completely detuning the second crystal and then turning the first crystal to an angle near the position for maximum 283-nm power using the dye laser stepper motors controlled by software (Sirah Control 3). The second crystal is then tuned by a piezo-driven micrometer screw (New Focus Picomotor) that allows for rotation of the second crystal independent of the first. Both crystals are enclosed by a housing fed with dry filtered air, with windows at Brewster’s angle for the incoming and exiting beams. With 175 W pumping from the Nd:YAG, the dye laser produced 56.8 W of 566-nm radiation and 7.0 W of 283-nm radiation after frequency doubling, yielding a dye conversion efficiency of 32% and a frequency conversion efficiency of 12%. The laser system operates continuously, using water cooling for both the Nd:YAG laser and the dye solution. Dye lifetime is on the order of hundreds of watt-hours, and can be preserved by the use of a variable attenuator between the pump and dye lasers, as implemented here. During the course of this experiment, we did not experience any loss of power due to dye depletion.

The 283-nm excitation beam is formed into a sheet using a plano-concave cylindrical lens and a plano-convex spherical lens. The vertically collimated sheet is 41 mm in height and converges to an approximately 0.3-mm waist at the probe volume. A fused-silica window reflects a small portion of the beam over a premixed CH4-air flame stabilized on a small test burner and then onto a photodiode fitted with a UV diffuser. OH laser-induced fluorescence is measured using a photomultiplier tube attached to a 0.1-m monochromator. This assembly allows for calibration and monitoring of both laser wavelength and laser pulse energy.
B. Imaging System

OH radicals are excited using the Q_1(7) transition of the A^2Σ^+→X^2Π (1,0) band, since the transition is strong and the ground state is suitably temperature insensitive. OH PLIF from the transitions in the A^2Σ^+→X^2Π (1,1) and (0,0) bands is recorded using a CMOS camera (Photron SA-5) with a two-stage image intensifier (LaVision HS-IRO) gated at 100 ns. Collection optics included a 45-mm, f/1.8 UV lens (Sodern Cerco) fitted with a high-transmission (> 80% at 310 nm) bandpass filter (Laser Components GmbH custom fabrication). Signal is digitized to 12-bits (equivalent to 4095 grey levels) and stored on the camera’s 32 GB of onboard RAM. For 50-kHz framing, the camera sensor’s active area must be reduced to 384 (H) × 344 (V) pixels, allowing for image sequences of greater than 173000 frames, or 3.46 s in duration. Flow features with a temporal frequency up to 25 kHz and as low as 0.3 Hz can be resolved, yielding a temporal dynamic range greater than 8.3 × 10^4. Of course, the minimum frequency and temporal dynamic range are proportional to, and limited only by, the amount of camera RAM, which can be increased to 64 GB for this camera model. The field of view is 46 (H) × 42 (V) mm, resulting in a resolution of 121 μm/pixel. For the results presented here, sequences of 1000 frames are sufficient for analysis. Some initial transient behavior of the imaging system was observed and was repeatable, where pixel intensity count increases from frame to frame before reaching steady-state, even for a steady uniform light source. Therefore, only the latter 500 frames were used in all analysis that involved averaging over a sequence. DaVis software is used to control camera image acquisition and equipment timing [124]. ImageJ was used for all image processing and analysis [125].

C. DC Plasmatron

A schematic of the transient-arc DC torch is presented in Figure 5.11. A high voltage is applied between the cathode and anode. The charge accumulates until the potential reaches a breakdown
voltage, and then the stored energy is discharged as a short-duration (~100 ns) arc propagates from the cathode towards the anode due to the gas flow. The process repeats with both non-completed glow discharges and arcing at intervals on the order of 100 μs [81, 82]. The plasmatron ignites a premixed CH₄-air flow and sustains the flame. The total flow rate is varied from 6 SLPM to 10 SLPM (gas velocity ~ 5 to 8 m/s) with a near-stoichiometric (slightly lean) CH₄-air mixture.

**Figure 5.11** Diagrams of the Hencken burner (left) and the transient-arc DC plasmatron (right).

### 5.3.2 Results and Discussion

#### A. Hencken Burner

Figure 5.12 contains a representative sequence of OH PLIF images from a flame stabilized above a Hencken burner. This stable laminar flame is used to evaluate the OH PLIF image quality as
the signal region area and uniformity are suitable for signal-to-noise ratio (SNR) calculations, whereas SNR of turbulent flames is difficult to estimate due to signal non-uniformity. The average SNR of an image sequence is calculated as

\[
SNR = \frac{1}{N} \sum_{i=1}^{N} \frac{\mu_i}{\sigma_i},
\]

(5.9)

where \(\mu_i\) is the mean signal and \(\sigma_i\) is the root mean square (RMS) noise of a 16 × 16 pixel region in frame \(i\) of a sequence with \(N\) frames. This method is expected to somewhat underestimate SNR as it assumes that the signal in the region is uniform and any variation is due to noise, while in truth even a small region in a stable flame will have variation in signal due to absorption, laser profile non-uniformity, and OH density variation.

Figure 5.12 Sequence of OH PLIF images of a laminar Hencken burner flame

Figure 5.13 contains a plotted signal profile and an image noting the locations of the regions used in the Hencken flame for SNR calculation. For a region centered 25 mm above the flame surface, where OH number density can be reasonably estimated as the equilibrium value of \(\sim 10^{16}\) cm\(^{-3}\), the mean value of \(\mu_i\) (\(\bar{\mu}\)) = 799 and SNR = 13.5 (standard deviation of 1.5). For a higher intensity region, located near the flame surface, \(\bar{\mu} = 3200\) and SNR = 20.3 (standard deviation of 2.16). These values can be improved by accounting for fixed pattern noise of the imaging
system. Each frame of the Hencken flame sequence was divided by the 500-frame ensemble average of a cold cathode light panel (Coherent) to reduce the contribution of fixed pattern noise. SNR improved to 15.1 for the region 25 mm above the flame surface, and 23.6 for the region close to the flame. A two-by-two pixel averaging yields further improvements in SNR, to 30.6 and 18.3, but reduces the resolution to 242 $\mu$m/pixel.

![Graph](image)

**Figure 5.13** Plot of OH PLIF signal profile for Hencken flame. The location of the profile is marked with a line on the inset single-shot OH PLIF image. The squares mark the locations of SNR evaluation.

**B. DC Plasmatron**

The transient-arc DC plasmatron is used to ignite and stabilize turbulent premixed flames for demonstration of the diagnostic capability for practical application. Sequences of OH PLIF images are presented in Figure 5.14. The top images are at 10 SLPM flow and 600 mA average current. The current is reduced to 200 mA in middle section, while the flow is reduced to 6 SLPM in the bottom section. These images are unaltered raw data, other than the false color
applied. Based on a previous study, peak OH number density is expected to be on the order of $10^{16}$ cm$^{-3}$ for all these flames [126]. OH fluorescence signal is high, spanning the entire 12-bit range with an intensifier gain of 65% to 70%. The limited resolution and steep OH gradients make SNR calculation difficult, as regions that could be assumed of uniform fluorescence are not of sufficient size. However, peak signal levels are comparable to the high-intensity region Hencken flame images using the same intensifier gain (68%), and therefore the SNR is expected to be $\sim 20$ for these peak signal regions. Figure 5.15 shows a plot of the signal profiles at different heights for a sample single image.

Figure 5.16 contains a sequence of 20 frames cropped to highlight the evolution of transient flow features. It is clear that a lower framing rate would fail to record continuous flow development. For example, a 10-kHz system would produce only one out of every five frames, insufficient even at this relatively low velocity. At 50 kHz, the signal level and temporal resolution are sufficient to observe transient flow features interacting with the flame surface.
Figure 5.14 Sequences of OH PLIF in DC Torch plasma assisted flame for a premixed flowrate and current level of 10 SLPM and 600 mA (first), 10 SLPM and 200 mA (second), 6 SLPM and 600 mA (third), respectively.
Figure 5.14 (cont.)

Figure 5.15  Plot of OH PLIF signal profile for DC torch plasma assisted flame. The locations of the profiles are marked on the inset single-shot OH PLIF image.
The diagnostic system can be evaluated with respect to the ability to resolve turbulent flames of interest in the combustion community. The system presented here acquires images with a spatial resolution of 121 μm and a temporal resolution of 20 μs. This diagnostic instrument is suitable for combustion events with characteristic times as short as 100 μs, such as ignition, flameholding in a high-speed combustor, and flame front dynamics in turbulent flames. Additionally, the system can be useful for imaging of events within shocktubes and other short duration wind tunnels. Continued improvements in temporal resolution can be expected, both by Nd:YAG pumped dye laser systems and pulse burst systems, but improvements in CMOS readout rates are needed to take full advantage of said improvements in the laser system. Fortunately, CMOS

\textbf{Figure 5.16} Sequence of OH PLIF images for DC torch. Full frame image is marked to show selected sub-region. Δt = 20 μs.
camera readout rates continue to increase, and current models (e.g., the Photron SA-Z) can digitize a $1024 \times 1024$ pixel array at 20 kHz and a $640 \times 600$ pixel array at 50 kHz.

5.3.3 Conclusion

The advent of high-speed diagnostic equipment, including kHz-rate CMOS cameras, image intensifiers and tunable lasers, has enabled signal collection at framing rates sufficient to observe transient features of reacting flows. This study has demonstrated OH planar laser-induced fluorescence (PLIF) at a continuous repetition rate of 50 kHz in a laminar premixed CH4/air flame and a turbulent plasmatron torch (also with CH4-air). The average signal-to-noise ratio for the peak OH PLIF signals was estimated to be $>20$ in the laminar Hencken burner flame, and the sequences of images from the DC plasmatron are of comparable signal level and thus fidelity. The spatiotemporal resolution is sufficient to clearly capture turbulent flow features in this jet flame, and record lengths of $>170,000$ images are possible. The current state of diode-pumped Nd:YAG and tunable dye lasers are thus clearly sufficient for 50-kHz OH PLIF measurements.
CHAPTER 6  NO PLIF

Nitric oxide is an important molecule in combustion in terms of its role as an indicative precursor of NOx products [127], as well as being a stable PLIF tracer for diagnostics of other thermodynamic parameters [128-131]. Over the years, PLIF imaging of NO has been extensively carried out for many laboratory flames and practical combustion systems [132-134]. The results discussed here provide the demonstration of high-frequency NO PLIF imaging as a diagnostic tool for optimization of both current and future propulsion and chemical-energy conversion systems.

6.1 10 kHz NO PLIF

This study demonstrates high-repetition rate planar laser-induced fluorescence (PLIF) imaging of both cold (~300 K) and hot (~2400 K) nitric oxide (NO) at a framing rate of 10 kHz. The laser system is composed of a frequency-doubled dye laser pumped by the third harmonic of a 10-kHz Nd:YAG laser to generate continuously pulsed laser radiation at 226 nm for excitation of NO. The LIF signal is detected using a high framerate, intensified CMOS camera, yielding a continuous cinematographic propagation of the NO plume where data acquisition duration is limited only by camera memory. The pulse energy of the beam is ~20 μJ with a spectral width ~0.15 cm⁻¹, though energies as high as 40 μJ were generated. Hot NO is generated by passing air through a DC transient-arc plasma torch that dissociates air. The plasma torch is also used to ignite and sustain a CH₄-air premixed flame. Cold NO is imaged from a 1% NO flow (buffered by nitrogen). The estimated signal-to-noise ratio (SNR) for the cold seeded flow and air plasma exceed 50 with expected NO concentrations of 6000 to 8000 parts per million (ppm, volume basis). Images show distinct, high-contrast boundaries. The plasma-assisted flame images have a SNR of less than 10 for concentrations reaching 1000 ppm. For many combustion applications,
the pulse energy is insufficient for PLIF measurements. However, the equipment and strategies herein could be applied to high-frequency line-imaging of NO at concentrations of 10 to 100 ppm. Generation of 226-nm radiation was also performed using sum-frequency mixing (SFM) of the 532-nm-pumped dye laser and 355-nm Nd:YAG third harmonic but was limited in energy to 14 μJ. Frequency tripling a 532-nm-pumped dye laser produced 226-nm radiation at energies comparable to the 355-nm pumping scheme.

6.1.1 Experimental Setup

A. Laser Diagnostics

Figure 6.1 diagrams the laser diagnostics setup. NO radicals were excited using the $A^2\Sigma^+-X^2\Pi (0,0)$ band, requiring UV radiation near $\lambda=226$ nm. The laser configuration is similar to the typical Nd:YAG-pumped tunable dye laser often utilized for PLIF studies based on a 10-Hz laser system, with high-frequency lasers and detectors replacing conventional components. A diode-pumped, Nd:YAG Q-switched laser (EdgeWave Innoslab IS12II-E) operating at 10 kHz produces 355-nm radiation using two LBO frequency-conversion crystals. A tunable dye laser (Sirah CREDO), designed for high-frequency, high-power operation, produced output at 452 nm using Coumarin 450 dye (Exciton) within both the oscillator and the single-stage amplifier. An integrated frequency-control unit houses a BBO crystal for frequency doubling to 226 nm; based on the laser specifications, a linewidth of ≤0.1 cm$^{-1}$ is expected at 452 nm (and ≤0.15 cm$^{-1}$ at 226 nm). A pyrometer measured the 355-nm dye laser pump beam at 33.7 W and the 226-nm beam at 380 mW, yielding pulse energies of 3.37 mJ and 38.0 μJ, respectively. Lenses shaped the 226-nm beam into a sheet approximately 35 mm in height and 0.5 mm in width at the probe volume. A mirror was temporarily placed in the beam path to steer the light into a static cell filled with
NO diluted in N₂, allowing for excitation scans using a monochromator fitted with collection optics and a photomultiplier tube for LIF detection.

![Figure 6.1](image)

**Figure 6.1** Experimental setup. A removable mirror can be inserted to direct the beam through a static cell for excitation scans

The dye lifetime was estimated to be 20 minutes at full pump power. The dye was replaced twice during the course of this experiment, once after installation and alignment of the lasers and again following the setup of the optics and camera. A beam dump was positioned in front of the dye laser during warm-up of the pump laser to avoid unnecessary decay of the dye. Fortunately, at 10 kHz image acquisition takes only a fraction of a second (to fill the camera’s 8 GB RAM buffer). It should be noted that it is the pump power that is at issue here vis-à-vis the dye lifetime, not the repetition rate. While the repetition rate is three orders of magnitude greater than with ‘conventional’ 10 Hz applications, the pump laser power is only one order of magnitude greater. It should be noted too that the water cooled resonator and amplifier dye reservoirs are each 4 L in
capacity and equipped with high flow rate pumps, having been designed for a pump laser power of up to 80 W. Finally, we note that while frequency doubling is the simplest approach (at least in terms of the optical configuration) to achieving the requisite wavelength (and thus the approach to be used first in this study), two alternatives, sum-frequency mixing (SFM) and dye-laser tripling, were explored to mitigate the challenge of a short dye lifetime commensurate with the use of a Coumarin dye. These alternatives will be discussed in more detail below.

Imaging equipment included a CMOS camera (Photron SA-5), capable of readout rates of about $7 \times 10^9$ pix/s, and image intensifier (Invisible Vision UVi) equipped with a 45-mm, f/1.8 UV lens (Cerco) fitted with a 235-nm long-pass filter and a 300-nm short-pass filter (Asahi Optics). The intensifier gate was $\Delta t_{\text{gate}} = 200$ ns. Image sizes were $768 \times 768$ pixels, with a field of view of about $35 \times 35$ mm at a resolution of $45 \times 45$ µm per pixel. A pulse generator (Berkeley Nucleonics Corporation 575) triggered the laser and detection systems at 10 kHz.

**B. Cold Seeded Flow**

Planar laser-induced fluorescence was recorded in an NO-seeded non-reacting flow. A mixture of 1% NO in N\textsubscript{2} exits an unpowered plasma torch (i.e., acting as a passive flow device) as a jet surrounded by air co-flow (see Figure 6.2). The NO-seeded flow was adjusted from 2 to 10 standard liters per minute (slpm) and the co-flow adjusted from 0 to 20 slpm, following the IUPAC definition of standard temperature and pressure as 273.15 K and 1 atm. This results in a jet exit velocity of 1.7 to 8.5 m/s and a co-flow velocity up to 57 m/s.
Figure 6.2 Cross-section of the transient-arc DC plasmatron (left), and photographs of an air-plasma discharge (top right) and a CH₄-air flame

C. Transient-Arc DC Plasmatron

The discharge system used in this study to generate hot NO is a transient-arc direct-current (DC) plasmatron [20, 81, 82] from Applied Plasma Technologies; a schematic is shown in Figure 6.2. This plasma torch operates on a repetitive glow-to-spark transition mode, offering the advantages of a thermal plasmatron with low average power output, low average temperatures (600–1300 K) [83], and high electron density. The torch operates primarily in the glow mode where high voltage (~10 kV) is applied between the cathode (inner electrode) and the surrounding anode. During the glow-to-spark transition, a short-duration spark (~100 ns) propagates from the cathode towards the torch tip. As the overall temperature is low, the electrode erosion is significantly less than with traditional DC torches, and the torch can produce stable operating conditions without internal cooling. The plasmatron exit has an inner diameter of 5 mm. Quantitative measurements of NO have been performed previously at 10 Hz acquisition
frequency using this plasmatron, with concentrations reported as 5000 to 15000 ppm in an air-
plasma and 500 to 3500 ppm in a plasma-assisted flame [83, 126].

There are two gas inlets in this discharge system: Feed 1 guides gas flow directly to the arc chamber, and Feed 2 provides co-flow around the main exit of the torch. An air-plasma is produced by flowing compressed dry air into Feed 1 at 6 slpm, yielding an exit velocity of 5 m/s, and applying a voltage potential between the electrodes. For plasma-assisted combustion, CH₄ is added to the feedstock gas (Feed 1) at the proper rate to yield a stoichiometric mixture at a total flow rate of 6 slpm, and the plasmatron ignites and sustains a flame. Air is directed through Feed 2 at 10 slpm, acting as a co-flow for the cold flow case. It is noted that the flowrates used in this study are too high for unassisted flame stabilization.

6.1.2 Results and Discussion

The excitation spectrum was obtained by scanning the dye laser wavelength as a photomultiplier tube and oscilloscope recorded the fluorescence signal from the static cell containing diluted NO. Figure 6.3 contains the recorded signal and a simulated excitation spectrum from LIFsim, a multilevel excitation and fluorescence model [135, 136]. Simulation parameters included a temperature of 298 K, pressure of 1 atm, and an excitation laser linewidth of 0.15 cm⁻¹. The high resolution of the recorded spectrum produces clearly identifiable NO transitions and is in good agreement with the LIFsim model. The cold NO-seeded flow was excited using 226.26-nm radiation, located in the Q₁ and P₂₁ bandhead, while the hot NO produced by the air-plasma and plasma-assisted flame was excited using the relatively temperature insensitive overlapping transitions at 226.03 nm, composed of the P₁(23.5), Q₁+P₂₁(14.5), and Q₂+R₁₂(20.5) lines. Both sets of transitions are free of interference from the Schumann-Runge bands of O₂.
Figure 6.3  Photograph of static cell and fluorescence-collection devices for excitation scanning (top), a plot of the recorded excitation scan with target transitions marked (center), and LIFsim simulation (bottom).
A. Cold Seeded Flow

Figure 6.4 contains image sequences for an N₂ flow with NO seeded at 1%. No image processing has been done, except two-by-two pixel binning resulting in a spatial resolution of 90 × 90 µm per pixel. The fluorescence signal is very high, as would be expected for such a large concentration of NO and low rates of electronic quenching (with a buffer gas of N₂). NO concentrations are estimated to peak around 8000 ppm due to mixing with the co-flow. The intensity is fairly uniform from shot to shot, with minimal variation due to laser power fluctuation. At a time interval of 0.1 ms between frames, the transient flow structures are clearly defined.

![Figure 6.4 Image sequences of NO PLIF in a NO-seeded flow at 5 and 10 slpm containing 1% NO.](image)

B. Air Plasma

Figure 6.5 contains image sequences of an air plasma discharge at two current settings. Again, the only processing of the raw data is a two-by-two software binning operation. Based on a
reduced electric field of 50 to 85 Td [83], defined as the ratio of the electric field to the concentration of neutral particles and measured in Townsend (Td, 1 Td \( \equiv \) \(10^{-17}\) V·cm\(^2\)), the majority of energy deposition into the air flow is expected to result in N\(_2\)-vibrational excitation along with some electron excitation (about 5%) [137]. The primary mechanism for NO production in this discharge is most likely N\(_2\) (v) + O \rightarrow NO + N [138]. Again, flow features are well defined, and the high frame-rate captures feature development. Increasing the current produces a more turbulent discharge with increased vorticity in the mixing layer.

Figure 6.5  Image sequences of NO PLIF in a DC plasma torch air-discharge at 400 and 800 mA.

C. Plasma-Coupled Flame

Figure 6.6 contains image sequences of a plasma-ignited and sustained CH\(_4\)-air flame. The images have been gamma adjusted for greater visibility, as well as software binned. The low signal level is immediately apparent, as the NO concentration decreases greatly compared to the air-only plasma, a finding previously observed in this system and others [83, 126, 139, 140], and the low signal levels produce a grainy texture throughout the recorded sequences. Images
captured without a laser pulse confirm the increasing nonuniformity of the intensifier as the gain is increased to capture weak fluorescence, with the signal peaking in the center of the image (and falling off towards the edge). NO concentration has been estimated at a maximum of 1000 ppm for premixed CH$_4$-air flames produced by the plasmatron. The addition of CH$_4$ introduces alternative electron-impact reactions and a quenching partner for vibrationally excited N$_2$, both of which decrease the rate of NO formation. Thermal NO formation is the most probable dominant mechanism, as temperature ranged from 1850 to 2600 K [127]. Fuel contributes to the elimination of NO through re-burn reactions and production of atomic oxygen and ozone that react with NO.

Figure 6.6 Image sequences of NO PLIF in a plasma-ignited and stabilized flame at 400 and 800 mA.
D. Image Quality

Successful planar imaging requires adequate signal-to-noise ratio (SNR), or profiles will be blurred and difficult to identify at best. We are presenting numerical measures of our recorded SNR for multiple image sequences. The SNR was estimated using the equation

\[
\text{SNR} = \frac{\mu_{\text{signal}}}{\sigma_{\text{RMS}}} \tag{6.10}
\]

where \(\mu_{\text{signal}}\) is the mean signal in a small region (about 0.5 \text{ mm}^2 \text{ or 250 pixels}), and \(\sigma_{\text{RMS}}\) is the root mean square variation. The interrogated region was located in a region with the highest signal and was chosen to have minimal expected variation in NO concentration. Therefore, the reported values are estimates of the greatest SNR that can be expected for a single image after examining numerous sequences of each flow. Figure 6.7 contains plots of the SNR for each flow under multiple conditions with the estimated NO concentration noted. The error bars show the range of expected SNR for any given image. Both the cold seeded flow and the air plasma have a high SNR, typically 50 to 65 for concentrations estimated at 6000 to 7000 ppm. A more thorough study may produce higher SNR values, especially for the cold NO flows, as high turbulence makes regions of uniform signal scarce (and, therefore, the choice of a region without signal variation difficult). Additionally, the fixed pattern noise of the imaging system was not accounted for in the data analysis. It should be noted that the fluorescence yield and SNR are high in this mixture of NO and N\(_2\), as is the contrast between the jet core and surrounding air (due in part to the strong electronic quenching by O\(_2\)). If the carrier gas were instead air, the SNR would be greatly reduced due to strong electronic quenching by O\(_2\), and the contrast between unmixed and mixed jet fluid would be substantially less.
Figure 6.7 Signal-to-noise ratio for each of the three flow types studied, estimated from image regions of greatest signal. Error bars describe the typical spread over multiple images.

In examining the plasma-enhanced flame, the SNR is estimated as 5 to 10, with NO concentrations less than 1000 ppm. The magnitude of noise and signal strength is demonstrated in Figure 6.8. A profile one pixel tall is plotted from a representative image from each flow type. For many combustion applications, NO concentration will be one to two orders of magnitude less than in the plasma-enhanced flame, and NO PLIF would require a more energetic laser beam. However, the beam energy used here (~20 μJ) would be well suited for high-frequency line imaging, where measurements could be performed for NO concentrations as low as 10 to 100 ppm. Line imaging may also be possible for two-photon absorption laser-induced fluorescence (TALIF) O-atom measurements at nearby wavelengths (225 to 226 nm) at good signal level with a properly sized beam. Furthermore, one could potentially employ a normal
CCD camera in a kinetic operation mode, where the intensifier gates repeatedly while the CCD shifts the line image repeatedly before readout.

![Figure 6.8](image.png)

**Figure 6.8** Signal profiles for a seeded flow, air-plasma discharge, and plasma-assisted flame with inset pictures marked to depict location of data extraction.

### 6.1.3 Alternative Methods of 226 nm Generation

Two alternative methods of obtaining 226 nm radiation were explored after image acquisition was completed: 1) SFM of beams from the Nd:YAG and dye lasers and 2) dye-laser tripling. Both approaches use dyes that have lifetimes estimated at 10 hours or more, depending, of course, on the exact Nd:YAG pump power. With the SFM approach, both the second and third harmonics from the Nd:YAG are employed: the second harmonic beam (at 532 nm for this Nd:YAG laser) pumps the dye laser (operated at 622 nm, using Exciton Rhodamine 640 laser dye) while the dye output is then frequency mixed within a BBO crystal with the third harmonic (355-nm) beam from the Nd:YAG. This approach is used commonly with conventional Nd:YAG-dye systems and is known to be very efficient; here, however, the results were somewhat disappointing with a derived 226-nm pulse energy of only 14 μJ. With the tripling approach, the dye laser, operated at 678 nm (using Exciton LDS 698 laser dye), is pumped by the
second harmonic beam from the Nd:YAG (with the tripling unit removed). Two BBO crystals were employed, for dye-laser frequency doubling and then tripling, along with a custom half-wave plate placed between the two crystals; the resulting 226-nm output was 34 μJ/pulse, which is comparable to the highest pulse energy achieved with the doubling approach using Coumarin dye.

6.1.4 Conclusion

As high repetition rate diagnostics mature, the scope of measurements and applications continues to expand. We have demonstrated NO PLIF at a rate of 10 kHz, exciting NO radicals using the third harmonic of a diode-pumped Nd:YAG laser (operating at 10 kHz) to pump a dye laser, which was frequency doubled to a final beam wavelength near 226 nm and a pulse energy of ~20 μJ. Detection was performed using a high frame-rate CMOS detector and intensifier. The laser wavelength was verified through fluorescence excitation scans of NO in a static cell. Characterization of two flows with high NO concentration (~8000 ppm), one produced by a DC transient-arc plasmatron using air feedstock and the other a cold NO-seeded flow, was successful, and cinematographic image sequences were produced. The high signal-to-noise ratio (SNR) in these sequences, typically 50 to 65, allows for clear observation of transient flow features, such as vortex shedding. The DC plasmatron was used to ignite and stabilize a CH₄-air flame, producing a flow of much lower NO concentration, at most 1000 ppm, and SNRs of 5 to 10. PLIF images capture the flamefront, but lower signal produces images that typically lack sharp contrast. Our observations suggest that the strategy demonstrated in this work is inadequate for NO PLIF of many combustion applications, where NO concentrations are one or two orders of magnitude lower than those imaged here. Nonetheless, kHz-rate line imaging is an option for concentrations as low as 10 to 100 ppm. Finally, alternative strategies using sum frequency
mixing (SFM) and dye laser frequency tripling were explored as a means of mitigating the short
dye lifetimes, but it was hoped too that a more energetic 226-nm beam might be produced. While
SFM produced a 226-nm pulse energy of 14 µJ, and was difficult to configure, dye laser
frequency tripling produced 34 µJ/pulse (and was much easier to configure). Clearly, then, the
tripling approach holds promise for future measurements.
A novel strategy for PLIF of the methylidyne (CH) radical at kHz-rates was explored. It is demonstrated that the \( C^2\Sigma^+ \rightarrow X^2\Pi \) band is a good option for high-speed CH PLIF due to its strength and the convenience of the excitation wavelength (\( \lambda = 314 \) nm) that can be generated by a dye laser operating with red dye. Within this band there is significant interference from hydroxyl (OH) fluorescence. However, this can be avoided by careful wavelength selection. The strength of the transition is very favorable for continuous kHz-rate laser systems which produce low pulse energies (\( E_p < 1 \) mJ). The method is first demonstrated using a 10-Hz system to demonstrate its potential. The study then moves to 10-kHz operation in laminar and turbulent CH\(_4\)-air flames.

### 7.1 CH PLIF via C-X Excitation

#### 7.1.1 Introduction

Imaging of combustion radicals using PLIF is a widely utilized and common combustion diagnostic. As demonstrated in the previous chapters, OH and NO imaging are attractive target species. OH PLIF in particular is often employed as a flamefront marker, through its long lifetime causes some ambiguity in complex flowfields. CH is another candidate for flame front imaging, and CH PLIF has been demonstrated in previous studies [141-144]. CH concentrations are lower than OH, but it has the advantage of a distribution that better corresponds to the peak heat release rate than OH [145]. There have been two primary approaches to CH PLIF. The first implementations used the \( A^2\Delta \rightarrow X^2\Pi \) (0,0) band (\( \lambda = 314 \) nm) for both excitation and detection. The advantages of using the \( B^2\Sigma \rightarrow X^2\Pi \) (0,0) band for excitation was demonstrated by Carter et al. [146] wherein the B-X emission is rejected while the electronic energy transfer from B (\( v=0 \))
to A (v=1) allows for emission to be collected from the A-X(1,1) transition [147]. This method only collects the A-X fluorescence, which is a fraction of the total fluorescence.

The CH PLIF examined here uses a novel technique of using the C^2Σ^+→X^2Π (v′=0,v″=0) [148-150] band near 314 nm where excitation and emission efficiencies are significantly larger than the traditional A^2Δ→X^2Π (0,0) and B^2Σ→X^2Π (0,0) transitions. While the C state has been reported to be strongly predissociated [151], Jeffries et al. noted that the fluorescence yield is not significantly affected at atmospheric pressures and above, as electronic quenching should be the dominant decay mechanism [149]. Moreover, the C−X(0,0) emission coefficient is large, ∼9 × 10^6 s⁻¹, as are the absorption coefficients; for example, for most Q-branch transitions, B₁₂ > 2.5 × 10¹⁰ m⁻² J⁻¹ s⁻¹ [152]. As a result, even with a 314-nm pulse energy of 0.1 mJ and a laser sheet size of 75 mm × 0.3 mm, the excitation rate with an 8-ns laser pulse is ∼10⁹ s⁻¹. This makes a C-X excitation strategy very attractive for low pulse energy laser systems, such as kHz-rate Nd:YAG pumped dye lasers that can make use of long-lived red dyes.

There are some potential disadvantages of a C-X method, and perhaps these are some reasons that using this transition has not been explored. The greatest of these is likely the need to perform on-resonance detection. The off-diagonal bands are very weak, and so the excitation wavelength is within the fluorescence collection spectral range. This may create hurdles for imaging of an enclosed combustor, but the strength of the transition permits low pulse energies to be used, mitigating scattering concerns somewhat.

### 7.1.2 Experimental

We conducted CH PLIF measurements using a 10-Hz based laser system (Spectra-Physics GCR-170 Nd:YAG laser and Lumonics HD300 dye laser) operating with DCM laser dye. The 314-nm
laser beam was expanded into a uniform 48-mm-high sheet that was focused over the flame. Horizontal polarization of the laser beam was maintained to minimize Rayleigh scattering interference. A fused silica window was placed in the beam path to sample a small portion of the beam using a fast photodiode for timing purposes. We detected fluorescence with an ICCD camera (Roper PIMAX-3) fitted with a 100-mm focal length, f/2.8 UV lens (Cerco). A plano-convex lens, coated for UV, was mounted onto the front of lens to reduce the required standoff distance to less than the lens minimum focal distance. Lastly, a Schott UG5 colored-glass filter was used to suppress chemiluminescence in the visible range.

The burner consisted of a central tube with an inner diameter of approximately 5 mm for the main flow, surrounded by concentric 150-mm diameter tube for coflow. The central tube issued a chemically pure CH4 and air mixture, and the surrounding coflow was air at a velocity of \( \bar{V}_{CF} \approx 0.26 \text{ m/s} \) based on a flowrate of 250 SLPM. To generate a stable laminar flame, the fuel flowrate was 0.352 SLPM while the air flowrate was set to 2.41 SLPM, producing a flame with an equivalence ratio \( \phi = 1.40 \).

\subsection{7.1.3 Results and discussion}

A concern with this approach for detecting CH is the potential for interference from OH \( A^{2 \Sigma^+} \rightarrow X^2 \Pi (0,0) \) transitions. To better understand potential interference, we conducted an excitation scan, using a 10-Hz-based laser system, employing a laminar Bunsen flame (at \( \phi = 1.4 \)). The spectrum was prepared by defining a region of interest (ROI, for averaging signal) that lay along the CH layer. In spite of the placement of the ROI, the strongest features are from the OH \( A \rightarrow X(0,0) \) band. Synthetic OH \( A \rightarrow X \) and CH \( C \rightarrow X(0,0) \) excitation spectra, generated using LIFBASE [152], are shown below the experimental plot in Figure 7.1.
Figure 7.1  CH C–X(0,0) excitation scans. Simultaneous OH and CH PLIF imaging can be obtained by wavelength modulation
Figure 7.2 Detailed CH C−X(0,0) excitation scan. Inset images correspond to the excitation spectrum at the arrows.

Figure 7.2 shows greater detail of the Q-branch with inset Bunsen flame images (from which the spectrum was derived) corresponding to specific spectral peaks. The strongest features are OH transitions from the P₁ branch—which are about 3× times stronger than the peak CH features at the ROI and 5× stronger for the peak OH LIF signal relative to the peak CH signal—but weaker OH lines from the A-X (1,1) band are also evident. The strongest CH feature, composed of overlapped Q₁(7), Q₂(3), Q₂(4) and Q₂(5) transitions, is at \( \lambda = 314.423 \text{ nm} \); this line is nearby the OH Q₁(6) transition of the A-X (1,1) band (at \( \lambda = 314.429 \)), and in the Bunsen flame images (Image D) one can see the some level of OH interference (albeit small). If the laser wavelength is
tuned to the adjacent blue-side CH peak (Image C), composed of the $Q_2(6)$ and $Q_2(2)$ transitions ($\lambda = 314.415$ nm), the interference is much reduced while the CH LIF signal is only moderately less; also, the peak to the blue side of this one (Image B), composed primarily of the CH C–X $Q_2(8)$ and $Q_1(9)$ transitions, is also well isolated. The next CH peak (Image A) is approaching the strong OH $P_1(12)$ line, and some OH interference is evident.

As noted above, the C–X (0,0) band is attractive due to the strength of its transition. Strong transitions, however, can lead to some degree of saturation, even with irradiance values generated with sheet illumination with our kHz laser system. While transition saturation can be useful, to reduce the sensitivity to quenching and sheet non-uniformity, here, it can lead to increased interference from the nearby OH transitions (and potentially more interference from other species too), depending on the degree of saturation and the exact excitation wavelength. With a sheet size of about 50 mm (high) × 0.2 mm (thick), a recommended maximum energy for excitation of the Q-branch transitions is 0.5 mJ; beyond this value, interference from OH LIF may be significant, due to partial saturation of the Q-branch CH transition.

7.1.4 Conclusion

We describe efforts to develop planar laser-induced fluorescence (PLIF) of the CH radical for application to premixed flames and low laser pulse energy. The basic approach used here involves excitation and detection of the CH radical via the $C^2\Sigma^+–X^2\Pi$ (0,0) band, which has transitions in the wavelength range $\lambda \approx 310-320$ nm. Transitions in this band are generally stronger than those in the A–X and B–X bands of CH and the radiative lifetimes are shorter too; thus, use of the C–X band should have advantages with regard to CH detectability in atmospheric flames, especially for high-repetition-rate excitation sources that emit low pulse
energies. Of course, strong OH lines, from the \( \Lambda^2 \Sigma^+ - X^2 \Pi \) (0,0) and (1,1) bands, lie nearby the strong CH Q-branch lines. While this can create some interference in the detection of CH, we show that OH lines can be avoided, if desired, or excited, if desired. Indeed, easy access to either CH or OH is a substantial benefit to the method outlined herein. Furthermore, we show that simultaneous excitation and detection of CH and OH is possible too.
7.2 kHz-rate CH PLIF

7.2.1 Background

This paper describes efforts to develop kHz-rate or high-speed planar laser-induced fluorescence (PLIF) of the CH radical. Our focus here is on application premixed CH₄-air flames, but the approach is general and can be applied to any flame (or reaction zone) wherein there is sufficient CH. The basic approach applied here was described in section 7.1 CH PLIF via C-X Excitation. Briefly, it involves excitation and detection of the CH radial via the C₂Σ⁺−X₂Π (v'=0, v''=0) band [148-150], which has transitions in the wavelength range λ≈ 310-320 nm. Transitions in this band are generally stronger than those in the A–X and B–X bands of CH, and the fluorescence lifetimes are shorter [152]; thus, the C–X band should have advantages with regard to CH detectability in atmospheric flames.

The hydroxyl radical (OH) is generally abundant in flames, from the flamefront to the products. This fact, combined with its relatively simple spectroscopy, strong A–X transitions, etc., makes it amenable to imaging with the PLIF, even with small pulse energies delivered by kHz-rate systems [51, 64]. Methylidyne (CH), on the other hand, is generally found in relatively small concentrations in flames, making its detection significantly more challenging, especially for imaging applications. Nonetheless, CH has been used as a flamefront marker since it exists only near the reaction zone [141, 143, 144, 146, 153]. Typically, CH has been excited using a low-repetition-rate laser system based on a 10-Hz Nd:YAG pump laser, using either the A-X or B-X bands, but other sources have been used too, including a frequency-doubled Alexandrite laser [154], a kHz-rate Nd:YAG-pumped dye laser [155], and an optical parametric oscillator (OPO) pumped by a pulse-burst laser [156] or a Nd:YAG cluster [157].
With regard to the advantages of the C–X band, the excitation wavelength is more convenient for dye lasers, than the A–X and B–X transitions, as it involves use of a long-lived red dye (e.g., Exciton DCM). For example, the wavelength for B–X excitation, ~390 nm, is most conveniently generated with a short-lived blue laser dye (Exciton Exalite), pumping the dye laser with the third harmonic of a Nd:YAG laser. On the other hand, the CH C–X excitation and fluorescence bands overlap (off-diagonal bands are very weak), and a UV collection lens is required. Fortunately, background scattering can be mitigated with the use of smaller pulse energy and fast UV camera lenses are now available, thus mitigating, at least to some degree, the aforesaid disadvantages.

### 7.2.2 Experimental Setup

Measurements with the 10-Hz system provided a baseline against which the performance of the kHz system could be judged (see section 7.1 CH PLIF via C-X Excitation). Both 10-Hz and kHz systems employed DCM laser dye. The kHz system consisted of an EdgeWave Nd:YAG laser (EdgeWave Innoslab IS12II-E), producing up to 62 W at 532 nm and operating at 10 kHz, and a Sirah Credo dye laser; the dye laser produced up to 14 W at 628 nm. An integral BBO crystal was used for frequency doubling of the dye laser beam, and powers up 2.2 Watts (0.22 mJ/pulse) were achieved near the peak of the dye curve.

The 314-nm beam was directed toward the flame using dielectric mirrors designed for a XeCl excimer laser. The PLIF sheet was formed using a -38-mm focal-length plano-concave cylindrical lens and a 750-mm focal-length spherical lens. The cylindrical lens produced a beam that was well expanded beyond the aperture of the focusing lens. The sheet was measured to be about 0.23 mm by scanning a 25-μm slit through the sheet and monitoring the transmitted beam.
Fluorescence was detected using a two-stage intensifier (LaVision HS-IRO, with gate time set to 100 ns) equipped with a Cerco 100-mm f/2.8 lens and coupled to a CMOS camera (Photron SA-Z). A Schott UG5 filter was used to pass the UV fluorescence and block visible emission (for example, from CH chemiluminescence). A UV close-up lens (rather than an extension ring), to work at distances closer than the minimum focal distance [158]. The setup is diagrammed in Figure 7.3.

The 2-D Bunsen burner employed for the turbulent flame measurements is described by Filatyev et al. [144] and Steinberg et al. [159], while the specific configuration was nearly identical to that described by Steinberg et al. Briefly, it consists of a central burner 25.4-mm in width by 50.8-mm in length. The central flame is anchored on either side by burners of identical dimensions operated at near stoichiometric conditions with mean reactant flow rate of about 0.37 m/s. We operated the central flame near stoichiometric conditions ($\phi = 1.07$) with flow rates corresponding to a bulk reactant flow rate of about 8 m/s. All gas flow rates were metered with Tylan electro-mechanical flow controllers. Purity of the CH$_4$ was 99%; the air was from a compressed-air source that was filtered for particles and compressor oil, but from previous use with particle image velocimetry (PIV), there were particles within the burner tube. The central burner was setup with a turbulence generating grid, located just upstream of a hexagonal matrix; in addition the burner tube, with length of ~230 mm, was filled about halfway with stainless steel beads (of 5.6-mm diam.). The resulting flame was conical and reasonably (though not completely) uniform in the burner’s depth dimension.
7.2.3 Results and Discussion

To quantify resolution with the kHz system, we imaged the CH layer from a laminar Bunsen flame (equivalence ratio $\phi = 1.05$). (Note that all image processing was performed with the ImageJ software package [125].) Here, the field of view (FOV) was $\sim 46 \times 46$ mm. Generally, the layer thickness (FWHM) was between 0.26 and 0.3 mm. Laminar flame simulations, using CHEMKIN II PREMIX and USC Mech II [104], predict a CH layer thickness that is much smaller, about 0.13 mm at $\phi = 1.1$ (with a peak concentration of about 10 ppm). Clearly our
resolution is limited by both the lens and intensifier. In the preceding 10-Hz study, the ICCD system imaged a CH layer thickness of about 0.2 mm for the same flame. The difference between the two systems is the blur imposed by the intensifier in each system. LaVision characterizes the resolution of each intensifier with a measurement of the modulation transfer function; the value given for this HS-IRO was 16 line-pairs/mm for a contrast of 5%, a value typical for HS-IROs. We also investigated the resolution for the “as coupled” intensifier-camera system. For this purpose, a focusing target was imaged in a 1:1 configuration with the Photron SA-Z CMOS camera only, using a Nikon 105-mm macro lens. The lens f-stop was set to 5.6, to ensure that blur due to any misalignment was minimal. Subsequently, the IRO was attached to the camera, the focusing between the IRO and camera was optimized, and the target was again imaged at 1:1. The original image was then modified with the application of a Gaussian blurring function (across the entire image), and this image was compared to the intensified image. The best match was obtained with an imposed Gaussian blur of 3 pixels full width at half maximum height (FWHM). This equates to a blur of ~0.14 mm for the kHz system.

A sample composite set of images from this burner is shown in Figure 7.4. Here, the bottom image was 5-mm downstream of the burner exit (at the bottom of the image); the middle image was 45 mm downstream, and the top image 85 mm downstream. Note that here and below, we employed the following image processing steps: i) an average background flame emission image was subtracted from all images; ii) the images were divided by a flatfield image that incorporated the laser intensity profile and the camera-intensifier response; iii) a 2-pixel-radius median filter was applied; and iv) the image pixels were binned 2 × 2 such that the resulting image size is 512 × 512 pixels. The median filter applies a mild amount of smoothing to the CH layer; other filtering schemes can be used too to help smooth the CH layers. For example, the “2D
Anisotropic Diffusion Filter” within ImageJ is particularly effective in smoothing the layers but also maintaining sharp gradients.

Figure 7.4 Composite CH PLIF image, from 3 locations above 2D turbulent Bunsen flame, at 5, 45, and 85 mm from the burner exit. The white lines show the border between images.
The images are of high quality and show well the effect of turbulence in wrinkling the flame surface. It can be seen too that in general the CH layers are relatively thin, and a comparison with the laminar Bunsen flame shows that on the whole the layers in the turbulent flame are not much thicker than the layer from the laminar flame; however, one can see thickened CH regions too. Furthermore, there are instances wherein a pocket appears within the reactants. Presumably, this sort of image results from 3D motion of the flame surface, and this portion of the flame is connected *out of plane* (and not an isolated pocket); of course, volumetric measurements would be needed to fully understand the shape of the flame surface and verify this hypothesis.

One can also see “fingers” of flame pinching off and then burning out within the product region. These features—pockets of products within the reactants and pockets of reactants within the products—were also seen in the work of Filatyev et al. {Filatyev, #949}, but of course there was no time resolution in that work. Scattering from particles is also evident in some fraction of the images, but because of the relatively low pulse energy (perhaps also due to the horizontal beam polarization), particle scattering is not particularly problematic. It is worth noting too that in general interferences are suppressed by virtue of the weak excitation (from low laser fluence levels) and strong Einstein A and B coefficients. An image montage of the flame is shown in Figure 7.5; this was recorded 45 mm from the exit of the burner. The montage, covering a time interval of 1.9 ms shows the time evolution and complexity of the flame surface.
Finally, an added advantage with this approach for CH detection is that one can also detect OH. Indeed, with only minor shifts in the laser wavelength, one can excite either the A–X (1,1)-band $Q_1(6)$ transition or the A–X (0,0)-band $P_1(12)$ transition. With the $Q_1(6)$ transition, signals are comparable to maximum CH PLIF signals (for a slightly rich Bunsen flame); exciting the $P_1(12)$ transition provides higher signals but more laser beam absorption too. Furthermore, calibration
of the CH LIF signal might be accomplished (of interest for quantifying NO production via the flamefront mechanism) by comparison of OH and CH LIF signals. Simultaneous excitation and detection of OH and CH can be accomplished too, and a sample image is shown in Figure 7.6. Here, the laser wavelength was tuned between the CH Q1(7) and OH Q1(6) transitions (λ≈ 314.425 nm), so that there was some OH signal visible but still with a well-defined CH layer. This approach can be useful, particularly with complicated flame topology, in distinguishing reactants from products. It can also serve the practical purpose of showing the extent of the pilot region, which should be composed of hot combustion products with visible OH signals (and no intrusion of cold gases).

### 7.2.4 Conclusion

We describe efforts to develop kHz-rate or high-speed planar laser-induced fluorescence (PLIF) of the CH radical for application to premixed flames. The basic approach used here involves excitation and detection of the CH radial via the C2Σ+–X2Π (v′=0, v″=0) band, which has transitions in the wavelength range λ ≈ 310-320 nm. Transitions in this band are generally stronger than those in the A–X and B–X bands of CH, and the radiative lifetimes are shorter too; thus, the C–X band has advantages with regard to CH detectability in atmospheric-pressure flames. Of course, strong OH lines, from the A2Σ+–X2Π (0,0) and (1,1) bands, lie nearby the CH C–X lines. While this can create some unwanted in the detection of CH, we show that OH lines can be avoided, if desired, or excited, if desired. Indeed, easy access to either CH or OH is a substantial benefit to the method outlined herein, and we show that simultaneous excitation and detection of CH and OH is possible too, a technique that may have broad utility. Of course, the PLIF technique, as implemented here, is a resonant one, and thus background scattering and/or OH fluorescence may be problematic. However, due to the use of small excitation energy, ~0.2
Figure 7.6 10-kHz combined CH-OH PLIF image from a premixed turbulent flame. The field of view is 46 mm × 46 mm.

mJ/pulse, from our kHz-laser system, background scattering was not a limiting factor (even though there were some particles in the flowfield), and we have shown that OH lines can be avoided too. Finally, we have shown the utility of this approach with 10-kHz rate CH PLIF and of combined CH and OH PLIF in a turbulent, premixed CH₄-air Bunsen flame. The acquired images, which are of high quality, show the dynamics of the flame: we see intrusion of flame and
products into the reactant-zone and fingers of reactants pinch off in the product-zone and then burn out.
CHAPTER 8 CONCLUSION

An investigation of new equipment, techniques, and applications of laser diagnostics was presented in this dissertation. This work was performed with the purpose of improving our capability to understand turbulent combustion physics. With recent advances in high-speed tunable laser systems and intensified CMOS camera technology, continuous cinematographic imaging of physicochemical parameters in reactive flows is quickly becoming a reliable diagnostics tool and kHz-rate PLIF has gained attention in the scientific community as a tool to make meaningful measurements of high-enthalpy flows, and aid in the design and evaluation of robust and reliable super- and hyper-sonic engines. Imaging of combustion radicals has been a cornerstone diagnostic of the field for the past two decades, allowing researchers to visualize flame structure and behavior, but has been limited to low-frequency snap shots. Increasing the frame rate and expanding pool of detectable species for high-speed planar-laser induced fluorescence (PLIF) has been the primary focus of this study, and the significant advancements in the state of the art in combustion diagnostics for the study of turbulent combustion have been reported.

The first integration of sustained high-frequency PLIF imaging in a supersonic combustor was executed as part of an experimental investigation of a supersonic cavity combustor. 10-kHz OH (PLIF) captured flame geometry and dynamics within a cavity flameholder in a supersonic crossflow (Mach 2). The combustor behavior has been examined for direct fuel injection into the cavity over a range of fuel flow rates., and while the frame rate is not sufficiently fast to resolve flame dynamics in general, these measurements allow the acquisition of a large and statistically significant database and represent a first step towards time resolution within a high-speed flowfield.
The maximum rate for OH PLIF using a continuously operating laser system (where the only limitation for data record length is the camera memory size) has been extended to 50 kHz. Results are presented for a stable Hencken burner flame and a turbulent flame produced by a transient-arc DC plasmatron. The signal-to-noise ratio has been demonstrated to be very good, and the data presented shows the combination of high-quality, fast, and long duration imaging achieved in the effort. This opens the door to integration into supersonic combustor studies. Of particular interest is the highly transient and not well understood ignition process that is critical for further advancements in propulsion.

Another expansion of diagnostic capabilities achieved in this work is PLIF imaging of both cold (~300 K) and hot (~2400 K) nitric oxide (NO) at a framing rate of 10 kHz. The laser system consisted of a frequency-doubled dye laser pumped by the third harmonic of a 10-kHz Nd:YAG laser to generate continuously pulsed laser radiation at 226 nm for excitation of NO. The LIF signal is detected using a high framerate, intensified CMOS camera, yielding a continuous cinematographic propagation of the NO plume where data acquisition duration is limited only by camera memory. The estimated signal-to-noise ratio (SNR) for the cold seeded flow and air plasma exceeds 50 with expected NO concentrations of 6000 to 8000 parts per million (ppm, volume basis) and Images show distinct, high-contrast boundaries. The plasma-assisted flame images have a SNR of less than 10 for concentrations near 1000 ppm. For many combustion applications, the pulse energy is insufficient for PLIF measurements. Generation of 226-nm radiation was also performed using sum-frequency mixing (SFM) of the 532-nm-pumped dye laser and 355-nm Nd:YAG third harmonic but was limited in energy to 14 μJ. Frequency tripling a 532-nm-pumped dye laser produced 226-nm radiation at energies comparable to the 355-nm pumping scheme. It is quite possible that this proves to be the best strategy for kHz-rate NO PLIF.
A novel strategy for PLIF of the methylidyne (CH) radical at kHz-rates was explored. It is demonstrated that the C-X(0,0) band is a good option for high-speed CH PLIF due to its strength and the convenience of the excitation wavelength (\(\lambda = 314\) nm) that can be generated by a dye laser operating with red dye. Within this band there is significant interference from hydroxyl (OH) fluorescence. However, this can be avoided by careful wavelength selection. The strength of the transition is very favorable for continuous kHz-rate laser systems which produce low pulse energies (\(E_p < 1\) mJ). The method is first demonstrated using a 10-Hz system to demonstrate its potential. The study also applies the method to 10-kHz operation in laminar and turbulent \(\text{CH}_4\)-air flames with much success. Additionally, the ability to perform combined OH-CH PLIF is demonstrated.

As part of the effort to improve combustion performance in high-Reynolds number flows, investigations of plasma enhanced combustion have been performed. A tunable microwave waveguide was used to initiate and enhance combustion by coupling an atmospheric plasma discharge to a premixed \(\text{CH}_4\)-air flame. The absorbed microwave power ranges from 60 to 150 W, generated from a continuous source operating at 2.45 GHz, whereas combustion power ranges from 200 to 1000 W. OH radical number densities were measured using planar laser-induced fluorescence (PLIF) and temperatures were measured using Rayleigh scattering thermometry for various flow rates, equivalence ratios, and power levels. Increases in reaction volume, OH density, and temperature were observed as power increased.

Premixed and non-premixed flames are studied, using both solid and hollow inner conductors in the plasma applicator nozzle. The maximum temperatures occurred in the non-premixed flame, where the plasma is generated in air, reaching values of 3500 K. Temperatures are lower, peaking at 2000 K, when the plasma is generated at the air-fuel boundary or the air-premixed boundary through use of the hollow inner conductor. Non-premixed configurations were found
to be ill-suited for flame enhancement, whereas a premixed flow through the hollow electrode best demonstrated non-thermal plasma assisted combustion.

A 10-kHz study was also performed using an Nd:YAG laser to pump a frequency-doubled, high-speed tunable dye laser to excite OH radicals in the plasma-enhanced flame. Fluorescence and chemiluminescence from a microwave-plasma-ignited and stabilized flame are simultaneously recorded by intensified CMOS detectors at 10,000 frames per second (fps) to yield new understanding of the plasma-assisted flameholding mechanism and allow tracking of individual flow features.
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