

**DETERMINATION OF FLAME CHARACTERISTICS IN  
A LOW SWIRL BURNER AT GAS TURBINE  
CONDITIONS THROUGH REACTION ZONE IMAGING**

A Dissertation  
Presented to  
The Academic Faculty

by

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In Partial Fulfillment  
of the Requirements for the Degree  
Doctor of Philosophy in the  
Guggenheim School of Aerospace Engineering

Georgia Institute of Technology  
December 2012

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## LIST OF SYMBOLS

$X_f$  Flame standoff distance

# CHAPTER 1

## EXPERIMENTAL METHODS AND CONSIDERATIONS

The current chapter describes the facilities and apparatus used to study the flame characteristics in a Low Swirl Burner. The selection and implementation of diagnostic techniques used in this study are explained, as are data analysis methods used to process the acquired data.

### 1.1 LSB configurations

Two configurations of the Low Swirl Burner were tested for this study. There are referred to in what follows as Configurations A and B. Each configuration consists of the reactant flow inlet, the swirler device, the conduit to the combustion zone and the combustion zone itself. All swirlers tested for this work have an outer diameter,  $d_s$  of 38 mm (1.5 in). Other key dimensions of the swirlers tested are presented in Table 1.1.

Each configuration is housed in a high pressure testing facility. The testing facility consists of an air and fuel supply system, a pressure vessel with adequate optical access and an exhaust system for the products. Each testing facility is instrumented to measure temperatures and pressures which are then used to calculate various flow parameters of interest.

The design of the configurations tested, along with that of their respective test facilities are discussed in greater detail in this section.

#### 1.1.1 Configuration A

Preliminary experiments involving velocity field mapping and flame imaging were performed using this configuration. The schematic of the high pressure test facility

Table 1.1: *The dimensions of the swirlers used and the respective perforated plates are presented. Each swirler is referred to by its vane angle (as in “ $S_{37^\circ}$ ”).*

Geometric parameter	Swirler	
	$S_{37^\circ}$	$S_{45^\circ}$
<b>Swirler data</b>		
Outer diameter, $d_s$ , mm	38	38
Diameter ratio, $\frac{d_i}{d_s}$	0.66	0.66
Vane angle, $\alpha$	$37^\circ$	$45^\circ$
Theoretical Swirl Number, $S$	0.48	0.64
<b>Perforated plate data</b>		
Open area, $\text{mm}^2$	155.97	156.98
Blockage, %	71.54	71.36
Plate thickness, mm	1.27	1.27
Hole pattern	1 - 8 - 16	1 - 8 - 16
Hole location (dia), mm	0 - 10.2 - 19.1	0 - 10.2 - 19.1
Hole diameter, mm	2.79 - 2.79 - 2.84	2.82 - 2.82 - 2.83

housing this configuration is shown in Figure FIXME, while the configuration itself  
is shown in greater detail in Figure FIXME.

#### 1.1.1.1 Test Facility

Pressurized air is supplied from external tanks and heated in an indirect, gas-fired  
heat exchanger to about 500 K. The flowrate of the air is metered using a sub-critical  
orifice flow meter with a 38 mm (1.5 in) bore diameter Flow-Lin orifice plate capa-  
ble of metering a maximum flow rate of 2.2 kg/s (1 lb/s). The orifice flow meter  
is instrumented with an Omega PX725A-1KGI pressure transmitter calibrated to a  
reduced pressure range of 0–2.758 MPa (0–400 psi), a shielded K-type thermocouple  
and an Omega PX771A-025GI differential pressure transmitter, calibrated to a re-  
duced differential pressure range of 0–68.948 kPa (0–10 psid). The fuel (natural gas)  
is metered using a similar set up as the air line, with a sub-critical orifice flow meter.  
The orifice plate is a Flow-Lin orifice plate with a bore diameter of 13.46 mm (0.53



in), capable of metering a maximum flow rate of 0.22 kg/s (0.1 lb/s). The upstream pressure is measured using an Omega PX725A-1KGI pressure transmitter (same as the air line) and the differential pressure is measured using a PX771A-100WDC differential pressure transmitter with a pressure range of 0–2.489 kPa (100 in H<sub>2</sub>O). The temperature of the fuel is assumed to be the same as the room temperature (300 K).

The air enters the inlet nozzle of the LSB through a 1.8 m (6 ft) long, 102 mm (4 in) diameter straight pipe section. The straight pipe section allows for the flow to be fully developed, and fully premixed before the reactants enter the burner. The combustor pressure and temperature are measured at the head of the inlet nozzle. The pressure is measured by an Omega PX181B-500G5V pressure transducer with a pressure range of 0–3.45 MPa (0–500 psi), while the temperature is measured using a K-type thermocouple.

The pressure and temperature measurements are used to calculate the four primary flow parameters (combustor pressure, preheat temperature, reference velocity and equivalence ratio) for the LSB in real time. All measurements are monitored and recorded during the course of the experiment by a LabView VI.

The pressure vessel enclosing the combustor is designed to withstand pressures of up to 30 atm and is insulated from the combustor by a ceramic liner. Cooling for the pressure vessel and the quartz tube is provided by a flow of cold air introduced at the head of the pressure vessel. The cold air is drawn from the same external tanks as the main air line, but bypassing the heating system. The cold air flow is not metered, but its upstream pressure is coupled to the main air line so as to ensure a steady flow of cold air into the pressure vessel at all operating conditions. Optical access to the combustor is provided through four 25 mm (1 in) thick, 150 mm (6 in) × 75 mm (3 in) quartz windows located 90° apart azimuthally. The view ports allow the combustor to be imaged from the dump plane to an axial distance of 150 mm (6 in) downstream.

The exhaust from the combustor is cooled by circulating cold water through a water jacket enclosing each section of the exhaust pipe. The length of the exhaust pipe sections is about 1.8 m (6 ft). The exhaust pipe section terminates in an orifice plug to provide the back pressure to the combustion chamber. Different diameter orifices are used for each reference velocity condition to be tested. The exiting products are finally released to the building exhaust system.

#### 1.1.1.2 Low Swirl Burner

The detail of the LSB configuration is shown in Figure FIXME. The premixed, preheated reactants reach the swirler through a converging nozzle that decreases linearly in diameter from from the inlet diameter of 102 mm (4 in) to the outer diameter of the swirler, 38 mm (1.5 in). At the swirler, the flow splits into two streams — one passing through the central section and another picking up swirl by flowing over the vanes in the annular region. The relative flow split between the two streams is controlled by inducing blockage into the central flow by means of a perforated plate. The swirler leads to a constant area nozzle, and is located one diameter upstream of an abrupt area change. At the area change, the reactants expand from the 38 mm (1.5 in) diameter nozzle into a 115 mm (4.5 in) diameter combustion zone. This expansion ratio is chosen so as to avoid confinement effects on the centerline flame flow field.[\[1\]](#)

The main combustion zone begins at the dump plane and is enclosed by a GE 214 quartz tube. The quartz tube is 300 mm (12 in) long and 115 mm (4.5 in) in diameter. The thickness of the quartz tube is 2.5 mm (0.1 in).

#### 1.1.2 Configuration B

This configuration was used to image the flame structure of the LSB flame using CH PLIF. A schematic of the flow system of the test facility is shown in Figure FIXME,

while the LSB combustor itself is shown in greater detail in Figure FIXME.

#### 1.1.2.1 Test Facility

This test facility shares the upstream supply of preheated air, cold air and fuel (natural gas) with the one used in Configuration A. The flow rate of the preheated air stream is measured using the same orifice flow meter system used in Configuration A — albeit with a smaller 27.59 mm (1.0863 in) diameter bore Flow-Lin orifice plate. The fuel flow rate is measured using a critical orifice with a diameter of 0.8128 mm (0.032 in), instrumented with a 0–FIXME pressure transmitter, a K-type thermocouple and a FIXME differential pressure transmitter with a range of 0–FIXME. A short distance after mixing with the fuel (natural gas), the preheated reactants split into two separate streams. Each reactant stream is metered using an identical set up of .

Further, each reactant stream is individually metered by separate orifice flow meters. This builds redundancy in the system, offering a double-check of all readings and verifies that there are no leaks in the flow system. Each orifice flow meter is equipped with a thermocouple, an upstream pressure transducer and a differential pressure transducer.

The cooling air for the co-flow is not metered.

All measurements are monitored and recorded by a LabView VI.

The pressure vessel is rated for pressures in excess of 30 atm and is insulated from the combustor by a flow of cold air. The cold air enters the pressure vessel through two inlet ports and passes through a layer of steel ball bearings which renders the flow uniform spatially. The pressure vessel has four viewports located 90° apart for optical access. Each viewport is covered by a 25 mm (1 in) thick, 178 mm (7 in) × 50 mm (2 in) quartz window. The LSB exit is located approximately halfway between the top and bottom edges of the window, allowing about 88.9 mm (3.5 in) of the combustion zone to be imaged through the window. Similar to Test Rig A, the

exhaust section is cooled by circulating cold water through an enclosing water jacket. 118  
An adjustable gate valve on the exhaust line provides the back pressure necessary to 119  
pressurize the combustor. The products are vented into the same building exhaust 120  
system as Configuration A. 121

#### 1.1.2.2 Low Swirl Burner 122

The design of this LSB configuration is presented in Figure FIXME. As described 124  
earlier, the reactants reach the swirler device through two separate streams. The 125  
core/central stream passes through plenum chamber which is filled with steel ball 126  
bearings before approaching the swirler through a smoothly contoured nozzle with 127  
a high contraction ratio. The annular stream reaches the swirler directly through a 128  
separate contoured nozzle. The contraction ratio is chosen to inhibit the formation 129  
of thick boundary layers in the reactant streams. The core stream passes through 130  
the central section of the swirler, while the annular stream picks up swirl by passing 131  
through the vanes of the swirler. 132

The swirler device leads to a constant area nozzle which is FIXME in length. 133  
Following this, the reactants expand into the combustion zone. 134

Unlike in Configuration A, there is no dump plane or quartz tube to provide 135  
confinement to the combustion zone. The co-flow of cold air provides insulation to 136  
the walls of the pressure vessel. Further, in this configuration, the annular flow is 137  
separately controlled from the central flow, which allows one to control the mass flow 138  
split directly. Finally, the level of turbulence in the central flow can be adjusted by 139  
use of a turbulence generator[2] located upstream in the plenum chamber. 140

## 1.2 Diagnostics

### 1.2.1 Laser Doppler Velocimetry

The velocity field of the LSB is mapped using a TSI 3-component LDV system. Three wavelengths (514 nm, 488 nm and 476 nm) are separated from the output of a 5 W Argon ion laser by an FBL-3 multicolor beam generator. The individual beams are split into two coherent beams which are then focused to intersect and produce interference fringes within an ellipsoidal measurement volume with dimensions of the order of 100  $\mu\text{m}$ . For this purpose, two transceiver probes are mounted 90° apart about the axis of the LSB. One transceiver probe focuses the 514 nm and 488 nm beams in planes perpendicular to each other, while the second probe focuses the 476 nm beams orthogonal to the other two beams. Particles in the flow field crossing the interference fringes scatter the laser light elastically and produce a sinusoidal signal whose frequency is proportional to the velocity of the particle. The transceiver probes collect this scattered light and each wavelength is detected separately by a PDM-1000-3 three-channel photodetector module. The output from the photodetector is processed by an FSA-3500-3 signal processor. The resulting three components of the particle/flow velocity are recorded by the FlowSizer software.

Since the airflow is very sparsely populated by particles, the flow needs to be artificially seeded to facilitate LDV measurements in a reasonable amount of time. The seeding particles to be used and their mean diameter are decided by the characteristics of the flow to be imaged.[3] Since the LSB flow field is a reacting one, the particles need to have high melting points. Further, the particles need to be small enough to follow the flow closely and large enough or reflective enough to scatter light efficiently in the measurement volume. Based on these requirements, commercially available alumina particles with a mean particle diameter of 5  $\mu\text{m}$  were chosen for this study. In order to uniformly seed the flow, a novel seeding generator was designed as

described in Appendix A. The seeding particles were introduced slightly upstream of the 1.8 m (6 ft) long straight pipe section in Test Rig A.

LDV data was only acquired at atmospheric pressure conditions. At high pressure conditions, the reacting LSB flow field produces sharp refractive index gradients that rapidly shift in the turbulent flow field. This causes strong beam steering effects making it very difficult for the laser beams to reliably intersect within such a small measurement volume. The long distance traveled by the beams in the test rig further exacerbated this problem, making LDV data nearly impossible to acquire at such conditions.

### 1.2.2 $CH^*$ chemiluminescence

The LSB flame is imaged using one of two 16-bit intensified CCD cameras — PI Acton 1024×256 or 512×512 pixels — with a 28 mm f/2.8 camera lens.  $CH^*$  chemiluminescence is filtered using a bandpass filter centered on 430 nm with a FWHM of 10 nm. At each operating condition, 100 instantaneous images are acquired with an exposure of 1 ms. An additional 100 instantaneous images are acquired with no flame and averaged to yield the background for correcting the flame images.

$CH^*$  chemiluminescence has several advantages over flame chemiluminescence from other radicals such as  $OH^*$ ,  $C_2^*$ , etc. First, the  $CH^*$  emission occurs around 430 nm and is less affected by blackbody radiation from the walls of the combustor compared to longer wavelength detection, e.g.,  $C_2^*$ , which emits around 514 nm. Second, the intensity of the chemiluminescence from  $CH^*$  is known to scale well with heat release in the combustor[4], unlike  $C_2^*$ . Third, the emitted light can be gathered with high quantum efficiency by the intensified CCD cameras used for this study. The quantum efficiency of the 18 mm Gen III HB filmless intensifier used by the 512×512 camera is about 45% at 430 nm, compared to about 10% at 310 nm, where  $OH^*$  chemiluminescence peaks.

The flame chemiluminescence images acquired are background-corrected and averaged. The resulting mean is the line-of-sight integrated, time-averaged image of the flame. Strictly speaking, this is not the same as a real average obtained from a long exposure image as the instantaneous images are obtained through a periodic sampling process and hence, are prone to statistical errors. However, the behaviour of the flame can be assumed to be sufficiently random that the mean obtained is adequately representative of the true average. Figure FIXME shows a typical mean  $\text{CH}^*$  chemiluminescence image prepared in this manner.

Even when background-corrected, the walls of the combustor are not at zero intensity in the average chemiluminescence image. This is particularly noticeable near the dump plane where there is no flame present and yet the walls are clearly illuminated. The source of this background illumination is mostly the chemiluminescence from the flame scattering off the combustor and pressure vessel walls. The contribution from blackbody radiation from the heated walls is less significant in the narrow wavelength range imaged. This is evident from images acquired immediately after a flame blowout which show the walls to be nearly dark.

The averaged chemiluminescence image allows us to measure the flame standoff distance by following the intensity profile along the centerline of the combustor. The intensity profile rises sharply when passing the flame standoff location. Thus, the flame standoff location can be ascertained by finding the inflection point in the intensity profile.

The profile of the average chemiluminescence intensity along the centerline of the sample case from Figure FIXME is shown in Figure FIXME, showing the flame standoff distance. The distance from the dump plane, measured in number of pixels on the image and scaled by the appropriate magnification factor yields the flame stand-

off distance,  $X_f$ . The determination of the flame standoff location by this method provides a suitable and deterministic means to locating the leading edge of the flame front.

The average image can be processed further to yield more spatially resolved information about the flame brush. Under the reasonable assumption that the average LSB flame is axially symmetric about the centerline of the combustor, a tomographic deconvolution technique called an Abel deconvolution[5] can be used to convert the line-of-sight integrated image to a radial map of chemiluminescence intensity. In effect, this shows the shape and structure of the average flame brush. The Abel deconvolution of the sample data from Figure FIXME is shown in Figure FIXME.

The Abel-deconvoluted image provides an relatively easy means to determining the angle of the flame brush. A straight line joining two points located at the center of the flame brush intersects the axis of the combustor at this angle. The angle of the flame is denoted by  $\theta_f$ .

Using the Abel deconvolution to study the flame brush suffers from two main drawbacks. First, the system of equations describing the Abel deconvolution is only valid as long as the entirety of the flame is visible. This is only satisfied in the initial region of the LSB where the diameter of the flame brush is smaller than the height of the optical viewport. At further downstream locations, the flame is not imaged in its entirety. This causes the spurious bright regions near the top of the window in Figure FIXME. The second limitation of the Abel deconvolution technique stems from the high incidence of errors along the centerline (where  $r \rightarrow 0$ ). Due to this, any study of the flame brush thickness at the flame stabilization point — a metric of considerable importance — is all but impossible using this tomographic technique.



The CH PLIF setup uses the frequency-doubled output of a Light Age PAL 101 alexandrite laser tuned to  $\lambda \approx 387.2$  nm. The design of the laser is shown schematically in Figure FIXME. The active medium is a 150 mm (6 in) long, 5 mm (0.197 in) diameter alexandrite rod. The rod is placed between two flashlamps within the resonator cavity formed by two spherical mirrors. A birefringent tuning mechanism is placed within the resonator to allow the user to select the frequency of the output beam. The tuning mechanism is coupled to a micrometer whose reading relates linearly to the output wavelength. The tuning mechanism allows the fundamental wavelength to be varied between 720–780 nm, with peak gain at about 755 nm. The resonator cavity also contains two Q-switches, which allow the laser to optionally operate in double-pulsed mode. For this study, however, only one Q-switch was used and the laser was operated in single-pulsed mode only.

The diameter of the fundamental beam exiting the output coupler is reduced by a collimating telescope. This is done in order to increase the efficiency of conversion of the frequency-doubling crystal. The second harmonic portion of the beam is separated from the fundamental by a dichroic mirror and exits the laser. The fundamental beam is terminated at a beam dump within the laser.

The alexandrite laser is capable of operating at frequencies of up to 15 Hz. Laser power is controlled primarily by varying the voltage applied to the flash lamps. When operating with a high flash lamp voltage, it is recommended that the frequency of pulsing be reduced to allow more time to dissipate the heat build up within the alexandrite rod. All experiments conducted as part of this study operated the laser at 10.0 Hz.

The linewidth of the fundamental beam is determined by the manufacturer to be 150 GHz at  $\lambda = 775$  nm. Assuming the spectral profile of the laser to be a Gaussian, the linewidth of the frequency-doubled beam can be determined. The Full Width at

Half Max (FWHM) of a Gaussian curve scales linearly with the standard deviation  
of the curve. When convoluted with itself, the new standard deviation is  $\sqrt{\sigma^2 + \sigma^2}$   
or  $\sqrt{2}$  times that of the original curve. Thus, the new linewidth is  $150 \times \sqrt{2} = 212$   
GHz or  $7.07 \text{ cm}^{-1}$ . In wavelengths, this represents a spread of about  $1.06 \text{ \AA}$ .

The near-UV beam exiting the laser is used to pump the R-bandhead of the CH  
 $B^2\Sigma^- \leftarrow X^2\Pi (0,0)$  system. This populates the  $A^2\Delta$  state through fast electronic  
energy transfer from the  $B^2\Sigma^-$  state. The resulting broadband fluorescence observed  
between  $\lambda = 420\text{--}440 \text{ nm}$  is due to the  $A^2\Delta \rightarrow X^2\Pi (1,1)$ ,  $A^2\Delta \rightarrow X^2\Pi (0,0)$  and,  
to a lesser degree, from the  $B^2\Sigma^- \rightarrow X^2\Pi (0,1)$  bands. The LIF signal is collected  
using an intensified PI Acton  $512 \times 512$  camera equipped with an 18 mm Gen III HB  
filmless intensifier with a quantum efficiency of about 45% in the  $420\text{--}440 \text{ nm}$  range.  
Elastic scattering from the laser beam is attenuated by a 3 mm thick GG 420 Schott  
Glass filter.

#### 1.2.3.1 Laminar Flame setup

Preliminary experiments to evaluate the CH PLIF technique are performed on a  
laminar flame. The choice of a laminar flame as the subject allows us to neglect  
effects of strain and turbulence on the flame. Further, laminar flames are more readily  
simulated by reaction kinetics packages like Chemkin with high fidelity.

These experiments are conducted on an laminar, methane-air flame stabilized on  
an unpiloted Bunsen burner with an inner diameter of 10.16 mm (0.4 in). The air  
flow rate was measured and regulated using a Dwyer rotameter with a range of 0–  
20 SCFH calibrated using a Ritter drum-type gas meter. The natural gas flow rate  
was metered using a Matheson FM 1050 602 rotameter with a range from 0–1230  
SCCM. This flowmeter was calibrated using a Sensidyne Gilibrator 2 bubble flow  
meter system.

### 1.2.3.2 Laser Wavelength Calibration

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As described earlier, the output wavelength of the PAL 101 alexandrite laser is controlled using a micrometer-coupled birefringent tuning mechanism. The wavelength of the laser beam varies linearly with the micrometer reading. Initially, the manufacturer-supplied calibration for the micrometer was found to be inaccurate. This required an experiment to calibrate the laser output wavelength against the micrometer reading in order to determine the slope and offset of the calibration curve accurately.

A schematic of this experiment is shown in Figure FIXME. The laser beam is glanced off a steel optical post and the scattered light is collected using a fiber-optic cable coupled to an Ocean Optics HR 2000 spectrometer. The spectrometer is pre-calibrated using 50 wavelengths in the 400–850 nm range from the output of a Neon discharge lamp source. The spectrometer is also intensity corrected over this range using a black body source. The estimated error in the resolution of the device is about 0.1 nm (1 Å).

The laser micrometer was traversed from 0.600 in to 0.626 in and back in steps of 0.001 in. The calibration was performed using at the fundamental wavelength of the laser. Each spectrum recorded is integrated over 512 ms and averaged over 10 such acquisitions. The background-corrected peak of the spectrum is then modeled as a Gaussian and the location of the center of the Gaussian waveform is recorded.

The results from this experiment are shown in Figure FIXME. The variation of the wavelength is verified to be linear against the micrometer reading. Further, there is little difference between the measurements taken while increasing and decreasing the micrometer position. This indicates that any effects of hysteresis in the micrometer position are minimal. A linear fit is applied to the points on the graph and the correct calibration equation is thus obtained.

An excitation scan is performed by tuning the output of the alexandrite laser from  $\lambda$  = 387.077 nm to 387.260 nm. This serves two purposes. First, it locates the optimal wavelength to excite the CH radicals that results in the highest fluorescence yield. Second, the variation of the signal intensity can be compared with simulated profiles from LIFBASE or other spectroscopic calculations and our estimation of the laser linewidth can be validated. Knowing the laser linewidth accurately is important for modeling the LIF signal. This aspect will be discussed in further detail later in this thesis/chapter FIXME.

A schematic of the excitation scan experiment is shown in Figure FIXME. The intensified PI Acton 512×512 camera described in Section 1.2.2 is used to image a premixed, laminar methane-air flame operating at close to stoichiometric conditions. The laminar flame is stabilized on the Bunsen burner described in Section 1.2.3.1. The alexandrite laser is operated at a power of 16 mJ/pulse in the second harmonic. The sheet forming optics consist of a +50 mm cylindrical lens and a +250 mm spherical lens placed 300 mm apart. The optics form the beam into a collimated sheet about 25 mm (1 in) tall, focused to a thickness on the order of 100  $\mu$ m at the flame location. The sheet passes through the center of the flame and the edges of the sheet are blocked by razor blades to prevent reflections from the burner from saturating the camera.

The induced fluorescence in the flame sheet is imaged perpendicularly by the intensified camera using an 85 mm f/1.8 Nikon AF Nikkor lens. This gives a magnification of approximately 62  $\mu$ m/pixel. The camera is triggered by the flash lamp sync signal from the laser system and the intensifier is gated over 300 ns, encompassing the 70 ns laser pulse. The long gate width gives the intensifier enough time to prepare to receive the fluorescence, preventing signal loss due to iris-ing. The gate width is still short enough that minimal flame chemiluminescence or ambient lighting is recorded

in the images. 100 instantaneous images are acquired for each excitation wavelength  
to acquire a good estimate of the mean fluorescence signal,  $\mu_{sig}$ .

Figure FIXME shows a sample CH PLIF image from this dataset. The images  
are background-corrected by subtracting the laser scattering (recorded without the  
flame). The fluorescence signal is calculated from these images using three alternate  
approaches.

In Method I, two “windows” are identified that include the straight sections of the  
laminar flame. The average fluorescence signal in each frame is calculated by taking  
the average of all the emitting pixels in the two windows. A pixel is designated as an  
emitting pixel if its intensity exceeds the standard deviation of a typical background  
pixel by at least a factor of five. The average of this value over all the frames is  
designated as the mean fluorescence signal,  $\mu_{sig}$ . In Method II, the intensity of the  
pixels is integrated over a straight line connecting the inner and outer edges of the  
flame. The straight line is chosen along the beam so that the beam intensity does  
not vary along the integration path. The integration is performed on the left and  
right arms of the flame, giving two readings per frame. The mean of these values over  
all the frames is recorded as the mean fluorescence signal,  $\mu_{sig}$ . In Method III, the  
midpoints of the straight lines from Method II are located and the average of their  
intensities, over all the frames is recorded as the mean fluorescence signal,  $\mu_{sig}$ . The  
regions of interest for each of these methods is highlighted in Figure FIXME.

The result of this investigation is shown in Figure FIXME. The calculated mean  
fluorescence signals from the three methods are plotted against a LIFBASE simulation  
of the absorption spectrum of the CH  $B-X$  transition. The profiles are appropriately  
scaled to match the LIFBASE simulation at the maximum value and at the minimum  
value. The LIFBASE simulation is performed for a thermalized system at 1800 K, at  
atmospheric pressure. Further, the instrument linewidth is specified to be the same  
as our estimate of the laser linewidth (1.06 Å).

The profiles of the calculated and scaled mean fluorescence signals are observed to agree extremely well with the LIFBASE simulation result. The discrepancies between the three methods is minimal.

The results indicate that the optimal excitation wavelength, corresponding to the highest mean fluorescence signal, is about 387.2 nm. For the rest of the experiments performed in this work, the laser is operated at this wavelength. The results also help verify that the calibration of the micrometer is accurate and the wavelengths are precisely adjustable. Finally, the results validate that our estimated laser linewidth, 1.06 Å, is accurate. This value can now be used in subsequent calculations of the LIF signal levels.

#### 1.2.3.4 Linearity test

The next step is to estimate the variation of the LIF signal as a function of the operating conditions. However, this function depends on which LIF regime we operate in. Hence, it is imperative that we verify the regime of operation before attempting to model the CH PLIF signal.

Under the assumption that the CH ground state population is not depleted by excitation or laser-induced chemical reactions, the LIF signal is linearly proportional to the laser intensity in the weak excitation limit. However, as the laser intensity is increased further, the LIF output is observed to saturate and plateau. This is called the strong excitation limit/saturation regime. In the weak excitation limit, the signal is a function of CH concentration and the rate of collisional quenching of the excited CH radicals. In the strong excitation limit, the signal depends only on the CH concentration is unaffected by the quenching of the excited CH species.

It is difficult to ensure that the CH system is saturated spatially, temporally and spectrally at the same time. Further, operating with high laser intensities may bleach the energy levels being excited by inducing chemical reactions that destroy the excited

CH radicals. Hence, it is preferred to operate in the linear regime.

For this experiment, the laser beam is directed at a steady, laminar, methane-air, Bunsen flame operating at a slightly rich stoichiometry. The 1 mm diameter beam is passed through an aperture, but no optics are used to refract the beam otherwise. Varying the intensity of the laser beam by changing the flash lamp voltage or the Q-switch timing is not preferred as either would alter the pulse-width of the beam. Instead, quartz disks and blocks are introduced into the beam to produce an intensity loss through reflection, scattering and absorption.

The flame is imaged with the PI Acton 512×512 intensified camera equipped with a 50 mm, f/1.8 AF Nikkor lens and a 3 mm thick GG 420 filter. The resolution of the set up was measured to be about 44  $\mu\text{m}/\text{pixel}$ . The laser power was varied from 10 mJ/pulse to 0.5 mJ/pulse in the manner described earlier. The LIF signal image was recorded over 150 accumulations. The corresponding laser scattering image was also recorded at each power for better estimating the background. The flame chemiluminescence was also recorded for the same purpose.

The average signal per pixel is plotted in arbitrary units against the laser intensity in Figure FIXME. The results indicate that the linearity of the LIF signal is valid for all laser intensities under 1 J/cm<sup>2</sup>.

## CHAPTER 2

423

### CH PLIF SIGNAL MODELING AND VALIDATION

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#### 2.1 Fluorescence Signal Intensity

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As described in Chapter FIXME 2, the excitation scheme used in this study produces 426  
fluorescence through a three-step process. First, the CH radicals in the ground state 427  
 $X^2\Pi, v = 0$  are excited by the incident radiation to the second electronically excited 428  
state  $B^2\Sigma^-, v = 0$ . This excitation occurs near the R-bandhead and targets the 429  
ground state CH radicals present in the rotational energy levels,  $N = 5$  through 9. 430  
The upper electronic state  $B^2\Sigma^-, v = 0$  is nearly degenerate with the  $A^2\Delta, v = 1$  431  
energy level. This leads to the population of the  $A^2\Delta, v = 0, 1$  energy levels due to 432  
collisional energy transfer. The resulting fluorescence collected is primarily the result 433  
of three spontaneous transitions —  $A \rightarrow X(1, 1)$ ,  $A \rightarrow X(0, 0)$  and  $B \rightarrow X(0, 1)$ . 434  
These transitions are shown in Figure FIXME. 435

The primary goal of this exercise of modeling the CH fluorescence signal intensity 436  
is to gage the feasibility of using CH PLIF to study various premixed flames, rather 437  
than to quantitatively calculate the amount of CH present in the flames. As such, 438  
we are more interested in the order of magnitude of the PLIF signal, rather than the 439  
absolute value of it. 440

The intensity of the CH fluorescence signal may be written as a function of the 441  
amount of CH radicals present in the excited state and the probability of spontaneous 442  
emission from said state. Symbolically, this may be written as shown in Equation 443  
**B.1.** 444

$$S = nVA \quad (2.1)$$



In Equation B.1,  $S$  is the total number of photons emitted per unit time,  $n$  is the number of excited CH radicals in a unit volume,  $V$  is the volume from which the signal is observed. The Einstein coefficient for spontaneous emission,  $A$  represents the probability of spontaneous emission between the two involved energy states. The predicted signal intensity represents the total number of photons emitted in all directions. In reality, only a fraction of these emitted photons will be recorded by the collection system. This fraction is a function of the experimental setup and depends on the collection angle, the efficiency of the optics and the detector used to record the signal. This fraction is left out because our objective is only to predict the relative variation in the signal between various premixed flames.

This formulation of the signal intensity implicitly makes the following assumptions.

1. The fluorescence emission is predicted at steady state.
2. The collection volume is optically thin and an emitted photon is not reabsorbed within the flame itself. This is a reasonable assumption to make, since the flame thickness and the thickness of the laser sheet are both typically quite small.

As described earlier, an accurate model of the CH system should involve five energy levels —  $X(0)$ ,  $B(0)$ ,  $A(1)$ ,  $A(0)$ , and  $X(1)$ <sup>1</sup>. Such a model would also have to account for collisional transfers between each of these levels, in addition to spontaneous and stimulated transitions. The mathematical solution quickly becomes complicated and tedious. Further, it would involve several rate coefficients that have not been measured in experiments done so far.

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<sup>1</sup>In this notation, the letter represents the electronic energy level and the number in the parentheses represents the vibrational quantum number of the energy level

## CHAPTER 3

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### LSB FLAME CHARACTERISTICS

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In Chapter 2, we introduced the salient features of the Low Swirl Burner (LSB) flow field and discussed the mechanisms by which the LSB flame is stabilized. Further, various characteristics of the LSB flame that can be measured from flame images were outlined. To recapitulate, these are the flame location, flame shape and the flame structure. The first two are quantified by the flame standoff distance,  $X_f$ , and the flame angle,  $\theta_f$ , respectively.

In the same chapter, we introduced the four flow parameters that describe an operating condition for the LSB — the combustor pressure,  $p$ , the preheat temperature,  $T$ , the mass-averaged inlet velocity (also called the reference velocity,  $U_0$ ), and the equivalence ratio of the premixed reactants,  $\phi$ . We further introduced a geometric parameter — the angle of the vanes of the swirler,  $\alpha$ , which affects the amount of swirl present in the flow field.

The LSB flame was studied over a range of operating conditions, and the effect of flow and geometric parameters on the reacting flow field were investigated. The results of these investigations are presented in this chapter.

#### 3.1 Effect of reference velocity

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In typical gas turbine applications, varying the loading on the engine does not affect the reference velocity. However, since the reference velocity is a design parameter, the effect it has on the flame characteristics has implications for the design of future LSB-based gas turbine engines.

One of the key objectives of this thesis is to investigate how the LSB flame stabilization operates at high pressure conditions. The simple model described in Chapter

FIXME 2 predicts a self-similar flow field for the LSB at all reference velocities. This implies that the reference velocity will have no discernible impact on the flame standoff distance. This result is desirable for gas turbine designers, since the flame location and shape can be assumed to be constant. Limited testing conducted in published works confirmed this behavior at atmospheric pressure conditions with no preheat.

In order to verify the validity of this model at high pressure conditions in the presence of substantial preheat, the LSB was operated at a pressure of 6 atm over a range of reference velocities from 10 m/s to 40 m/s. For these tests, the  $S_{37^\circ}$  swirler was used. In a parallel series of tests, the  $S_{45^\circ}$  swirler was tested at a pressure of 3 atm at a reference velocities of 40 and 80 m/s. The location of the flame was measured from CH\* chemiluminescence images, and the results are presented in Figure FIXME.

There is essentially no systematic variation in the flame standoff distance or the flame angle for the low velocity,  $S_{37^\circ}$  tests. Based on the model, this can be interpreted as the increase in reference velocity producing a concomitant increase in the turbulent flame speed at the flame stabilization location, negating any change in the flame's location. In other words, the flow field appears to retain its self-similarity, even at elevated pressures and temperatures.

When the  $S_{45^\circ}$  swirler was tested at higher reference velocities, however, the flame location shifted downstream sharply. This indicates potential limitations to the simple flame stabilization model that may not predict the behavior of the LSB flame at elevated pressures and temperatures, particularly at high reference velocities.

A possible cause of this limitation can be explored by considering the effect of increased reference velocity on the turbulent combustion regime in which the LSB combustor operates. Previous studies have primarily operated the LSB in the flamelet regime where the modified Damköhler model predicts the behavior of the turbulent flame speed with reasonable fidelity. At elevated pressures, both the laminar flame speed of the reactants,  $S_L$  and the flame thickness,  $\delta_f$  are diminished. This places

the operating regime higher and more to the right on a Borghi diagram, as shown in Figure FIXME. While increasing the reference velocity did not affect the turbulent combustion regime at lower pressures in a flamelet combustion regime, at elevated pressures the flame may be transitioning into the thin reaction zone regime. This transition would cause a reduction in  $S_T/S_L$ , or at least a lesser increase, and the turbulent flame speed would no longer be expected to increase in step with  $U_0$  and the increased levels of turbulence. This would explain the the observed downstream shift of the high pressure LSB flame at high reference velocities.

### 3.2 Effect of preheat temperature

The preheat temperature of the reactants is a key flow parameter, especially for gas turbine combustors. In general, the rates of most chemical reactions in the flame zone are highly sensitive to the temperature of the reactants. For the LSB in particular, the temperature of the incoming flow directly affects its viscosity and consequently, the velocity field in the flame stabilization region. Thus, studying the effect of the preheat temperature on the LSB flame and flow field is important.

In order to explore this in greater detail, the velocity field of the combustor was mapped using Laser Doppler Velocimetry (LDV). The conditions were chosen to study the effect of increasing the preheat temperature on both reacting and non-reacting LSB flow fields. Further, the study includes both low and high reference velocity cases. The relevant flow parameters relating to these tests are presented in Table FIXME. All LDV tests were limited to atmospheric pressure conditions. Implementing the LDV technique at elevated pressures proved difficult due to beam steering issues, coupled with impractical turn-around times between the successive runs that would be required to obtain sufficient LDV data points for analysis.

The normalized centerline mean and rms axial velocity profiles for the three cases are presented in Figure FIXME. The abscissa represents the distance from a point

called the virtual origin,  $X_0$ . The virtual origin is defined as the imaginary location 543  
where the extrapolated linear axial velocity profile reaches the reference velocity in 544  
magnitude. The extrapolation is indicated in Figure FIXME by a dashed line. 545

As noted in Chapter FIXME 2, previous studies[6] reported that mean axial 546  
stretch — the normalized slope of the linear decay of axial velocity — at the in- 547  
let of the combustor was self-similar, regardless of the Reynolds number,  $Re$  of the 548  
operating condition. Further, it was reported that the velocity decay was steeper for 549  
reacting cases compared to non-reacting cases. 550

The results presented in Figure FIXME however, show that even though Cases 1 551  
and 2 have similar  $Re$ , their mean velocity profiles have very different slopes. Further, 552  
the reacting and non-reacting cases (both at preheated conditions) have similarly 553  
steep slopes. This indicates that the mean axial stretch in the near field of the LSB 554  
flow field is a stronger function of the preheat temperature than  $Re$ . The presence 555  
of preheat results in increased viscosity that enhances the momentum transport in 556  
the radial direction. This causes the velocity decay to be steep for preheated cases, 557  
compared to cases without preheat. 558

Assuming that  $S_T$  is constant, these results suggest that at higher preheat tem- 559  
peratures, the flame would stabilize closer to the dump plane because of the faster 560  
velocity decay and reduced local flow velocities. In fact, a faster velocity decay would 561  
produce greater  $u'$  values and increase  $S_T$ , further causing the flame location to shift 562  
upstream. Furthermore, in view of the steep velocity profile, it may be anticipated 563  
that any changes in the stabilization location caused by perturbations in the local 564  
flow field (and hence, the local turbulent flame propagation velocity) are likely to be 565  
of diminished magnitude in the presence of preheat. All of this leads to an intuitive 566  
result — the LSB flame behaves more stably at high preheat conditions. 567

### 3.3 Effect of swirler vane angle

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As described in Chapter FIXME 3, the LSB swirlers tested for this study are designed to have the same mass flow splits. The  $S_{45^\circ}$  swirler has a higher vane angle, resulting in greater blockage to the flow passing through the annular section. In order to compensate for this, the perforated plate covering the central section has slightly smaller holes. The net effect retains the same mass flow split as in the  $S_{37^\circ}$  swirler.

Chapter FIXME 2 describes how the swirler vane angle relates to the amount of swirl imparted to the incoming flow. According to Equation FIXME, a swirler with a higher vane angle will produce greater swirl in the reactants. Previous work in swirl combustion[7, 8] has pointed out that increased swirl shortens the flame by enhancing the swirl-induced radial pressure gradients. The data acquired in the present investigation is in agreement with this observation. Operated at identical inlet conditions, the  $S_{45^\circ}$  swirler stabilizes a flame closer to the dump plane and with a larger flame angle compared to the  $S_{37^\circ}$  swirler.

This result highlights an interesting trade-off for the designers of LSB-based gas turbine engines. The  $S_{45^\circ}$  flame is located further upstream and has a more concentrated region of heat release. This enhances the strength of the toroidal recirculation zone near the dump plane, which may be powerful enough under certain conditions (as we shall see in Section 3.4) to even cause the flame to attach itself to the lip of the inlet. All of this means that the  $S_{45^\circ}$  flame is more stable and will resist perturbations in the incoming flow better than the  $S_{37^\circ}$  flame. However, the presence of a strong recirculation zone in the flow field of the  $S_{45^\circ}$  swirler will entrain more hot products and retain them longer near the zone of heat release. This is a recipe for the production of thermal  $\text{NO}_x$ . While no emission measurements were made as part of this study, it may be reasonably anticipated that the  $\text{NO}_x$  performance of the  $S_{45^\circ}$  swirler will be degraded compared to the  $S_{37^\circ}$  swirler. The trade-off for gas turbine

engine designers is thus between flame stability and emissions performance.

### 3.4 Effect of equivalence ratio

The LSB is primarily intended for fuel-lean operation in order to utilize its low  $\text{NO}_x$  emission performance. As a result, most of the testing was done as close as possible to a target  $\phi$  of 0.56. Limited testing was carried out at 12 atm for two off-target conditions: a slightly richer ( $\phi \approx 0.58$ ) and a slightly leaner ( $\phi \approx 0.53$ ) mixture, in order to explore the sensitivity of the LSB flame to limited changes in equivalence ratio. The  $S_{45^\circ}$  swirler was used for these tests. The corresponding averaged and Abel-deconvoluted flame images are presented in Figure FIXME.

Two characteristics of the flame are immediately obvious from these images. First, the zone of heat release, marked by the region from which  $\text{CH}^*$  chemiluminescence is observed, is increasingly compact at fuel-rich conditions. Virtually all other flame images acquired at a leaner condition show a long flame, with the heat release distributed over the entire visible area of the combustor. The compactness of the heat release zone indicates potentially poor  $\text{NO}_x$  performance at these conditions.

Second, the fuel-rich flame brush can be observed to wrap around and anchor itself on the dump plane. This is particularly observable in the Abel-deconvoluted image. The attached region is not as bright as the rest of the flame brush, indicating that the flame may be attaching itself intermittently. This intermittent behavior can be confirmed from the instantaneous images where it is visible on some of the acquired images, but not others. This behavior was alluded to in Section 3.3 as being the result of the enhanced toroidal recirculation zone produced by this swirler. Thus, the intermittent attachment of the flame to the inlet indicates the increased importance of the toroidal recirculation zone in stabilizing the flame.

It should be noted that the reliance on a toroidal recirculation zone to anchor the flame to the inlet is one of the primary flame stabilization mechanisms used by

traditional swirl combustors. Thus, LSB swirlers with high vane angles tend to behave  
like traditional swirl combustors at fuel-rich conditions.

### 3.5 Effect of combustor pressure

In many gas turbine engines, the combustor pressure varies directly with the loading  
of the engine. Like the preheat temperature, the combustor pressure affects the LSB  
flame both through the fluid mechanics of the flow and the kinetics of the chemical  
reactions in the flame. The effect of the combustor pressure on the fluid mechanics  
of the LSB flow field can be captured by its effect on the Reynolds number. As noted  
in Section 3.2, however, previous work indicated the Reynolds number may not be  
an important parameter for the LSB, particularly in the near field where the flame  
stabilization occurs. On the other hand, the effect of the combustor pressure on reac-  
tion rates in the flame is clearly important. Increasing the combustor pressure results  
in a lower laminar flame speed and reduced flame thickness for methane-air flames.  
According to the modified Damköhler model discussed earlier, the reduced laminar  
flame speed should have little or no effect on the flow field, since the contribution from  
 $S_L$  in Equation FIXME is vanishingly small, even at the lowest reference velocities  
of our test conditions. However, as suggested by our discussion in Section 3.1, the  
validity of the simple model at elevated pressure conditions is questionable.

In order to resolve the uncertainties regarding how the LSB flame responds to  
combustor pressure, the flame was imaged over a range of operating conditions from  
3 to 12 atm. For these tests, the reference velocity and the equivalence ratio were  
held constant. However, the temperature of the reactants continues to increase with  
pressure. The reason for this was discussed in Chapter FIXME 3 and is attributable  
to the reduced heat losses in the connecting pipes at the high flow rates required to  
pressurize the LSB. The flame location and shape inferred from the flame images are  
presented in Figure FIXME.



At low to moderate pressures, the flame location is nearly invariant for  $S_{37^\circ}$ , but  
moves upstream for the  $S_{45^\circ}$  cases. This behavior can be explained as follows. The  
flame stabilization location for the  $S_{45^\circ}$  swirler is closer to the dump plane compared  
to the  $S_{37^\circ}$  swirler. This should result in enhanced heat transfer to the dump plane  
and consequently to the incoming reactants. This feedback is even more effective as  
the temperature of the incoming reactants increases. This causes the upstream shift  
of the  $S_{45^\circ}$  flame, while the  $S_{37^\circ}$  flame is less affected by these processes.

At high pressures, however, both flames are observed to move downstream, despite  
the increasing preheat temperatures. The apparent decrease in the turbulent flame  
speed at these conditions is an unexpected result, and the modified Damköhler model  
is insufficient in accounting for this observation. Figure FIXME also shows that the  
flame angle for both cases decreases slightly with pressure. This suggests that the  
turbulent flame speed was consistently decreasing with pressure. In light of this,  
the nearly constant location of the  $S_{37^\circ}$  flame could be attributed to the effects of  
increasing combustor pressure and preheat temperature nearly canceling each other  
out at the lower pressures.

### 3.6 Flame structure

## APPENDIX A

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### SEEDER DESIGN

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A new seeder was designed for use in high pressure implementations of diagnostic techniques like Laser Doppler Velocimetry (LDV), Particle Image Velocimetry (PIV), etc.

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The previous design, as shown in Figure FIXME, was a fluidized bed seeding generator. Seeding particles in a cylindrical vessel are fluidized by an air-turbine vibrator. Air is introduced into the vessel in the form of two opposing jets directed tangentially to produce a small amount of swirl in the flow field. Particles are picked up by the air flow and the swirl aids in separating the heavy/coagulated clumps of seeding particles by centrifugal acceleration.

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This design had several shortcomings. First, it is observed that the seeding density of the seeded flow generally decreases over time, even if the seeding particles have not been depleted. The seeding particles tend to coagulate over time, due to the buildup of moisture, static charge, etc. In such cases, the vibrator can no longer effectively fluidize the particles. Further, the tangential introduction of the air flow preferentially depletes particles near the walls of the container, leaving the center relatively undisturbed. The cumulative effect of these phenomena diminishes the effectiveness of the seeder.

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Second, the fluidized bed requires a minimum amount of seeding particles to function effectively. This requires the seeder to be refilled even before all the seeding particles are consumed.

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Third, when designed for high pressure applications, the seeder will become quite heavy due to flanges and other fittings. Such a setup cannot be easily fluidized using a reasonable-sized air-turbine vibrator.

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The new seeder design is shown in Figure FIXME, and resembles a funnel with a 688  
swirler located halfway up the stem. A perforated base plate holds the swirler and the 689  
seeding particles in the conical section of the swirler. Due to the steep angle of the 690  
sides of the conical section, the seeding particles continuously collapse into the central 691  
section. This negates any need for vibrating the system. Air is introduced from the 692  
bottom of the seeder and enters the vessel by passing through the swirler. Since all 693  
the air enters this way, there is a considerable amount of swirl in the resulting flow 694  
field, Heavy/coagulated seeding particles are flung outward, while lighter particles are 695  
carried with the air. After a sufficient distance to allow for the cyclonic separation 696  
to be effective, the seeded air passes through another perforated plate which further 697  
limits the presence of large clumps of particles. The exiting air is now spatially and 698  
temporally uniformly seeded. 699

## APPENDIX B

### CH PLIF QUENCHING MODEL

In order to calculate the intensity of the quenched CH PLIF signal in a flame, an improved model of the CH system was constructed and analyzed. According to this new model, CH radicals from the  $X$  ground state are excited to the  $B(0)$  upper state. This is followed by collisional transfer to the  $A(1)$  and  $A(0)$  states. The transfer between the nearly degenerate  $A(1)$  and  $B(0)$  states is partially reversible. The transfer between  $B(0)$  and  $A(0)$  is not reversible. This is followed by spontaneous emission as CH radicals transition from the  $A$  states to the  $X$  state. This results in a pseudo-three-level model as shown in Figure FIXME.

Figure FIXME indicates the rates of the various processes discussed. The subscripts 0, 1 and 2 represent the electronic energy levels  $X$ ,  $A$  and  $B$  respectively. Processes involving the  $A(0)$  state are differentiated from those involving the  $A(1)$  state by a prime ( $'$ ). With the exception of the nearly degenerate  $A(1)$  and  $B(0)$  states, most collisional excitation steps are neglected due to their low probability.

In this formulation, the signal intensity of the CH PLIF emission is given by Equation B.1.

$$S = (n_1 A_{10} + n'_1 A'_{10} + n_2 A_{20})V \quad (\text{B.1})$$

The spontaneous emission coefficients,  $A_{10}$ ,  $A'_{10}$  and  $A_{20}$  are obtained from various published papers[9, 10, 11]. The values used for this analysis are presented in Table B.1.

Equations B.2–B.4 describe the time variation of the number density of CH radicals in each excited state.

Table B.1: *The coefficients of spontaneous emission for transitions in the CH system are provided.*

Transition	Symbol	A, s <sup>-1</sup>
$B \rightarrow X(0, 0)$	$A_{20}$	$2.963 \times 10^6$
$A \rightarrow X(1, 1)$	$A_{10}$	$1.676 \times 10^6$
$A \rightarrow X(0, 0)$	$A'_{10}$	$1.832 \times 10^6$

$$\frac{dn_1}{dt} = -(A_{10} + Q_{10} + R_{12})n_1 + R_{21}n_2 \quad (\text{B.2})$$

$$\frac{dn'_1}{dt} = -(A'_{10} + Q'_{10})n'_1 + R'_{21}n_2 \quad (\text{B.3})$$

$$\frac{dn_2}{dt} = W_{02}n_0 + R_{12}n_1 - (A_{20} + Q_{20} + R_{21} + R'_{21})n_2 \quad (\text{B.4})$$

At steady state, the rate of change of the number density is minimal. Under this assumption, the LHS of Equations B.2–B.4 can be set to zero. This results in a closed set of linear equations in terms of the populations of the upper states. This set of equations is presented in Equation B.5.

$$\begin{bmatrix} A_{10} + Q_{10} + R_{12} & 0 & -R_{21} \\ 0 & A'_{10} + Q'_{10} & -R'_{21} \\ -R_{12} & 0 & A_{20} + Q_{20} + R_{21} + R'_{21} \end{bmatrix} \begin{bmatrix} n_1 \\ n'_1 \\ n_2 \end{bmatrix} = \begin{bmatrix} 0 \\ 0 \\ W_{02}n_0 \end{bmatrix} \quad (\text{B.5})$$

The solution to Equation B.5 is shown in Equations B.6–B.8.

$$n_1 = \frac{R_{21}}{(A_{10} + Q_{10} + R_{12})(A_{20} + Q_{20} + R_{21} + R'_{21}) - R_{12}R_{21}} W_{02}n_0 \quad (\text{B.6})$$

$$n'_1 = \frac{(A_{10} + Q_{10} + R_{12})R'_{21}}{(A'_{10} + Q'_{10})((A_{10} + Q_{10} + R_{12})(A_{20} + Q_{20} + R_{21} + R'_{21}) - R_{12}R_{21})} W_{02}n_0 \quad (\text{B.7})$$

$$n_2 = \frac{(A_{10} + Q_{10} + R_{12})}{(A_{10} + Q_{10} + R_{12})(A_{20} + Q_{20} + R_{21} + R'_{21}) - R_{12}R_{21}} W_{02}n_0 \quad (\text{B.8})$$

These expressions can be further simplified by noting various observations made 727  
in studies of the CH system. For instance, previous work[12, 13] has reported that 728  
the  $B$  state is slightly (about 1.3 times) more prone to quenching compared to the  $A$  729  
state. We can thus make the following assumptions. 730

$$Q_{10} = Q'_{10} = Q \quad (\text{B.9})$$

$$Q_{20} = 1.3Q \quad (\text{B.10})$$

Next, it has been reported[14] that the electronic energy transfer rate from  $B$  to 731  
 $A$  state accounts for 0.24 times the total collisional removal from the  $B$  state. 732

$$\frac{R_{21} + R'_{21} - R_{12}}{Q_{20} + R_{21} + R'_{21} - R_{12}} = 0.24 \quad (\text{B.11})$$

$$\therefore \frac{R_{21} + R'_{21} - R_{12}}{Q} = 0.4105 \quad (\text{B.12})$$

We further know[13, 14] that the collisional transfer from the  $B(0)$  energy level 733  
populates the nearly degenerate  $A(1)$  level about four times faster than the  $A(0)$  level. 734

$$\frac{R_{21} - R_{12}}{R'_{21}} = 4 \quad (\text{B.13})$$

Finally, it was observed[13] that the rate of forward transfer from  $B(0)$  to  $A(1)$  is about 1.6 times the reverse process.

$$\frac{R_{21}}{R_{12}} = 1.6 \quad (\text{B.14})$$

Collating Equations B.12–B.14, we obtain a closed set of linear equations. This can be solved to eliminate  $R_{21}$ ,  $R_{12}$  and  $R'_{21}$  in terms of  $Q$  as shown in Equation B.15.

$$\begin{bmatrix} R_{21} \\ R'_{21} \\ R_{12} \end{bmatrix} = \begin{bmatrix} 5.1966 \\ 0.4872 \\ 3.2479 \end{bmatrix} Q \quad (\text{B.15})$$

Substituting Equations B.9, B.10 and B.15 into Equations B.6–B.7 leads to simplified expressions for the populations of the upper electronic states purely as a function of the respective Einstein coefficients and the collisional quenching rate. These are presented in the following Equations B.16–B.18.

$$n_1 = \frac{5.1966Q}{(A_{10} + 4.2479Q)(A_{20} + 6.9838Q) - 16.8780Q} W_{02} n_0 \quad (\text{B.16})$$

$$n'_1 = \frac{0.4872Q(A_{10} + 4.2479Q)}{(A'_{10} + Q)((A_{10} + 4.2479Q)(A_{20} + 6.9838Q) - 16.8780Q)} W_{02} n_0 \quad (\text{B.17})$$

$$n_2 = \frac{(A_{10} + 4.2479Q)}{(A_{10} + 4.2479Q)(A_{20} + 6.9838Q) - 16.8780Q} W_{02} n_0 \quad (\text{B.18})$$

The quenching rate,  $Q$  of excited CH radicals is calculated by using the quenching cross-sections of various species. The quenching cross-sections are measures of the effectiveness of each collision between a given species and an excited CH radical. The effectiveness of the collision also depends on the velocity of collision between the two species,  $g_j$  and the abundance of the species,  $n_j$ . This relationship is formalized in Equation B.19.

$$Q = \sum_j g_j \sigma_j n_j$$

$$Q = \sum_j \sqrt{\frac{8kT}{\pi\mu_j}} \sigma_j \frac{pN_A}{RT} X_j \quad (\text{B.19})$$

In Equation B.19,  $\mu_j$  represents the reduced mass of the colliding CH- $j$  molecules,  $p$  is the pressure,  $N_A$  is Avogadro's Number,  $R$  is the Universal Gas Constant,  $T$  is the temperature, and  $X_j$  is the mole fraction of species  $j$ . The mole fractions of the various species in the flame, as well as the temperature across the flame are obtained from Chemkin simulations. The expression for the reduced mass is given in Equation B.20.

$$\mu_j = \frac{m_j m_{CH}}{m_j + m_{CH}} \quad (\text{B.20})$$

The quenching cross-sections of various species are obtained from various published papers [15, 16, 17] and are functions of temperature. The functional forms used in this study are presented in Table B.2.

The term  $W_{02}n_0$  in Equations B.16–B.18 represents the rate of pumping of the ground state CH radicals. The current excitation scheme targets multiple transitions in the R-bandhead. The pumping rate for each transition is the product of the number of CH radicals present in the appropriate level, the Einstein absorption coefficient for that energy level,  $B_i$  and the amount of laser energy available at the appropriate frequency,  $E_i$ . As a result, the term is actually a summation over the individual energy levels. Equation B.21 presents this symbolically.



Table B.2: *The functional form of the quenching cross-sections of various species with CH are provided.*

Species	$\sigma, \text{\AA}^2$
H <sub>2</sub>	$6.1 \exp(-686/T)$
H	$221T^{-0.5} \exp(-686/T)$
O <sub>2</sub>	$8.61 \times 10^{-6} T^{1.64} \exp(867/T)$
OH	$221T^{-0.5} \exp(-686/T)$
H <sub>2</sub> O	9.6
CH <sub>4</sub>	$52.8T^{-0.5} \exp(-84/T)$
CO	8.31
CO <sub>2</sub>	$8.67 \times 10^{-13} T^{3.8} \exp(854/T)$
C <sub>2</sub> H <sub>6</sub>	13.4
N <sub>2</sub>	$1.53 \times 10^{-4} T^{1.23} \exp(-522.1/T)$
C <sub>3</sub> H <sub>8</sub>	22

$$\begin{aligned}
 W_{02}n_0 &= \sum_i B_i I_i n_i \\
 W_{02}n_0 &= \sum_i B_i \frac{E_i}{A_c} \frac{p N_A X_{CH}}{RT} f_i
 \end{aligned} \tag{B.21}$$

Table B.3 presents the values of  $B_i$  for the transitions targeted by the current 765  
excitation scheme.[18] Assuming a Gaussian line shape for the laser, and using the line 766  
strengths from LIFBASE, the relative amount of energy absorbed by each transition 767  
can be calculated. These values are also presented in Table B.3. 768

In Equation B.21,  $A_c$  is the area of cross-section of the laser beam and  $f_i$  is the 769  
Boltzmann fraction of the population at the energy level  $i$ . The expression for the 770  
Boltzmann fraction at the energy level corresponding to the vibrational quantum 771  
number  $v$  and rotational quantum number  $J$  is given in Equation B.22. 772

$$f(v, J) = \frac{\exp\left(\frac{-hcE_v(v)}{kT}\right) (2J+1) \exp\left(\frac{-hcE_r(v, J)}{kT}\right)}{Q_{rv}} \tag{B.22}$$

Table B.3: *The coefficients of absorption for selected transitions in the CH X( $v = 0$ ) system are provided.*

$N''$	$\lambda$ , nm	$B$ , $\text{m}^2\text{J}^{-1}\text{s}^{-1}$	$E$ (normalized)
R1			
5	387.2698	$7.677 \times 10^9$	0.0568
6	387.1899	$7.665 \times 10^9$	0.1706
7	387.1677	$7.610 \times 10^9$	0.1483
8	387.206	$7.519 \times 10^9$	0.1479
9	387.308	$7.397 \times 10^9$	0.0126
R2			
5	387.2289	$7.539 \times 10^9$	0.1080
6	387.1549	$7.569 \times 10^9$	0.1128
7	387.1371	$7.539 \times 10^9$	0.0841
8	387.1786	$7.464 \times 10^9$	0.1311
9	387.283	$7.354 \times 10^9$	0.0279

The vibrational energy,  $E_v(v)$  of a level is calculated according to Equation B.23, 773  
while the rotational energy,  $E_r(v, J)$  is calculated according to Equation B.24. 774

$$E_v(v) = \omega_e \left(v + \frac{1}{2}\right) - \omega_e x_e \left(v + \frac{1}{2}\right)^2 + \omega_e y_e \left(v + \frac{1}{2}\right)^3 - \omega_e z_e \left(v + \frac{1}{2}\right)^4 \quad (\text{B.23})$$

$$E_r(v, J) = \left\{ B_e - \alpha_e \left(v + \frac{1}{2}\right) \right\} J(J+1) - \left\{ D_e + \beta_e \left(v + \frac{1}{2}\right) \right\} J^2(J+1)^2 \quad (\text{B.24})$$

The spectroscopic constants in Equations B.23 and B.24 are found in literature[19] 775  
and are provided here in Table B.4. 776

The rovibrational partition function,  $Q_{rv}$  is a summation over all available vibra- 777  
tional and rotational levels in the particular electronic state. For the ground state 778  
of the CH molecule, there are five available vibrational quantum numbers,  $v = 0$  to 779  
 $v = 4$ . The CH system falls under Hund's Case b and hence, the appropriate rota- 780  
tional quantum number to use is  $N$ . Each vibrational level has twenty-two possible 781  
values for  $N$  from  $N = 1$  to  $N = 22$ . For each rotational quantum number  $N$ , there 782  
are two possible values of  $J$  given by  $N \pm \frac{1}{2}$ . 783

Table B.4: *Spectroscopic constants for the CH X<sup>2</sup>Π level are presented.*

Constant	Value, cm <sup>-1</sup>
$\omega_e$	2860.7508
$\omega_e x_e$	64.4387
$\omega_e y_e$	0.36345
$\omega_e z_e$	$-1.5378 \times 10^{-2}$
$B_e$	14.459883
$\alpha_e$	0.536541
$D_e$	$1.47436 \times 10^{-3}$
$\beta_e$	$-2.530 \times 10^{-5}$

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