Laser-Induced Incandescence of Soot at High Pressures

by

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A thesis submitted in conformity with the requirements for the degree of Master of Applied Science
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University of Toronto

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Abstract

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Measurements of soot emission properties are of interest in both fundamental research and combustion-based industries. Laser-induced incandescence of soot particles is a novel technique that allows unobtrusive measurements of both soot volume fraction and particulate size with significant advantages. An apparatus utilizing this technique has been customized and used to provide measurements of soot concentration and particle sizing of a laminar, diffusion methane/air flame at pressures of 10, 20 and 40 atm at 6 mm above the burner. Soot volume fraction measurements correlate well with literature findings at all pressures. Despite similar trends, particle size values are found to be consistently larger than values reported in literature. Discussion on the errors of laser-induced incandescence as well as recommendations for improving the apparatus and results are herein.
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Chapter 1

Motivation

Recent studies have shown the direct impact of combustion-generated particles on global and local climate change [1]. Black carbon aerosols contribute to global warming by either direct radiative forcing or by reducing the surface albedo of portions of the planet [2]. Emission of soot also shows low thermal efficiency and its accumulation on the interior of internal combustion engines can have adverse effects on the engine operation leading to maintenance and reliability problems [3]. Intrinsically toxic, soot has also been shown to be detrimental to the human health and lead to exacerbation of pulmonary diseases [3–5]. On the other hand, the formation of these particles can be desired for efficient furnace operation and formation of carbon black [6–9]. Thus, understanding the underlying theory and finding suitable measurement techniques of combustion parameters are a must for establishing and enforcing policies and regulations in favor of the human’s health and environmental risks [6, 9].

Over the past 30 years, laser-induced incandescence (LII) has become an established and widely-used technique for measuring soot particle size and concentration due to its unique advantages over other techniques [6, 10]. Essentially, LII involves the laser heating of an absorbing particle to a temperature below its sublimation temperature using a high-energy pulsed laser with the subsequent photodetection of the incandescence emitted by
Chapter 1. Motivation

the particle [7, 11]. The magnitude of the incandescence signal determines the soot volume fraction whereas the decay rate of the signal is related to the size of the cooling particles [12, 13].

While traditional techniques are capable of measuring soot volume fraction through laser extinction [14], and primary particle diameter and aggregate size through laser scattering [14] and thermophoretic sampling/transmission electron microscopy analysis (TS/TEM) [15], these methods have various limitations [16]. The robust and unambiguous nature of LII, along with its conceptual simplicity and ease of implementation, has led to its success in providing high temporal and spatial resolution data. These data are used to describe both soot volume fraction and primary particle size in a wide range of applications such as laminar and turbulent flames, in-cylinder combustion, engine exhaust gas characterization [17] and pool fires [18].

Although LII seems to be quite well understood in atmospheric-pressure systems, accurate findings of pressure effects on the LII process remain unknown [10]. Additionally, even though the majority of practical combustion devices operate at elevated pressures, the current understanding of the influence of pressure on soot formation and oxidation is quite limited as well [3]. Very few researchers have studied the application of LII at elevated pressures [9, 10, 19, 20]. Hofmann et al. have studied the influence of pressure on the LII process [10] and also obtained soot primary particle sizes using experiments on a premixed flat ethylene/air flame at 1-10 bar [19], whereas Thomson et al. have measured soot concentrations and primary particle diameters using LII in a laminar co-annular non-premixed methane/air flame at pressures between 0.5-4.0 MPa [9]. In addition, Charwath et al. have investigated the constraints of two-colour TiRe-LII (Time Resolved-Laser Induced Incandescence) at elevated pressures [20]. Considering the lack of knowledge in this area, it is hoped that the presented results in this thesis will shed some light on combustion diagnostics at elevated pressures.
Chapter 1. Motivation

Objective

The objective of this thesis is to ready an LII apparatus for the investigation of flames within a high pressure combustion chamber and investigate the effects of pressure on soot formation parameters in methane/air flames.

This thesis builds upon the groundwork of Trevor Kempthorne [21] and Daniel D.E. Cormier [22] who completed extensive literature survey and assembled a functioning LII apparatus at atmospheric pressure. This work is to address the problems preventing the successful implementation of the apparatus at elevated pressures. Recommendations for future improvements on this project are to be given herein.
Chapter 2

Introduction

2.1 Alternative Measurement Techniques and Advantages of LII

LII is not the lone method of determining either particle size or concentration. Techniques such as laser attenuation, filtered Rayleigh scattering and transmission electron microscopy (TEM) are also frequently employed in place or in complement of LII. Laser attenuation is a technique used for measuring soot volume fractions. In this method, the ratio of absorbed to transmitted incident light is correlated to the soot concentration present in the flame [9]. However, in the case of high particulate concentration or the existence of turbulent conditions with high-frequency pressure and density fluctuations, accuracy of results obtained by this method decreases [7, 23].

Filtered Rayleigh scattering, in turn, measures the fraction of incident light that was scattered from molecules within a flame. The scattered light, upon calibration of the system, is used to provide particle temperatures [7, pp. 155-165]. When particulate concentration becomes large, this model fails and a mechanism, namely the Mie scattering, takes over, drowning out the weak Rayleigh signal. In this case, distinguishing between the existence of many small particles and fewer large particles becomes difficult.
A reliable physical technique for measuring particle size and distribution is looking at the soot with a microscope. In this technique (TEM), a microscopic grating is mechanically inserted into the flame of interest for microsecond durations to collect samples. One of the major problems with this method is the intrusive nature of physically inserting the grating into the flame, which noticeably alters the surrounding flame properties. Despite being labourious and time-consuming, this method is often used as a benchmark for more robust and non-intrusive methods such as LII [24, 25].

These techniques, although simple and capable of high accuracy measurements, are often difficult to apply to practical combustion conditions. An advantage of LII is that it relies on incandescence which is not a property unique to soot. Therefore, it can be used to detect other nano-sized particle concentrations as well. Also, since this method does not noticeably alter the behavior or properties of the flame, its non-intrusive nature allows real-time and instantaneous readings of both particulate concentration and diameter, instead of just time-averaged results. Aside from its sensitivity, accuracy, and versatility, the LII method is able to provide high temporal and spatial resolution data. The high temporal resolution results from the sampling rate of the detection equipments which is usually on the order of picoseconds. It is this quality that fits LII for in situ measurements of turbulent targets. The spatial resolution with LII is diffraction-limited and can be changed within or between experiments by simply changing apertures. In addition to all other advantages, LII has a large dynamic range of detection and has been used to detect particulate concentrations ranging from 0.01 ppt to 10 ppm in a single experiment [26]. In summary, LII is a robust method, which can be used from very high to very low pressures and concentrations of particulate, to give accurate results with a high dynamic range [9, 13, 27–29].
2.2 Laser Induced Incandescence Background

Initial interests in using LII as a soot diagnostics method were motivated when an LII signal was first detected as a source of interference in the application of spontaneous laser Raman diagnostics to soot laden combustion environments. Subsequently, techniques were placed on reasonably firm theoretical grounds regarding the potential of LII for soot volume fraction and particle size measurements. Finally, with the further development of the LII process, groups of researchers published papers addressing quantitative measurements for soot volume fraction. Based on the modeling of the particle-size-dependent energy transfer process from the heated soot aggregates and with modifications to the existing setups, LII finally became capable of yielding results for primary particle sizes [7].

Generally, there are two different ways of obtaining LII data: spatial LII and Temporally-Resolved LII (TR-LII). The former, which has been the primary method used by most researchers in the past, involves using a CCD camera to obtain snap-shot data of the entire flame. Using this method, position-dependent data is collected from the flame to obtain soot concentration measurements. The latter, which enables time-dependent data to be obtained, uses a slightly different equipment package and allows the experimenter to record the entire lifetime of the LII signal.

The major advantage of TR-LII is that it allows both the primary particle size and the soot volume fraction to be measured. Fundamentally, the magnitude of the LII signal determines the soot concentration whereas the temporal decay curve of the signal is associated with the primary particle size [12, 13]. The full theory as to how this is accomplished is discussed in the next chapter.

2.3 High Pressure Studies

In addition to great uncertainty regarding soot formation processes and soot properties, the control and prediction of soot levels in practical combustion devices are further
impeded by the complexities imposed by transient operating conditions, nonhomogeneous flow fields and other parameters which have a non-linear effect on soot formation [20, 30]. One of the most important parameters is pressure, which has a significant effect on soot formation and oxidation in combustion systems [3]. Although most practical combustion devices used in transportation systems and stationary gas turbine combustors operate at elevated pressures, our understanding of the effects of pressure on soot formation and combustion processes is limited.

Very few LII researchers have actually attempted to measure soot particle parameters in very high pressures [9, 10]. Generally, the application of high pressures in LII measurements faces many challenges in both experimental implementation and the interpretation of the detected signals. These challenges include faster temporal decays of the LII signal, higher soot sublimation temperature, beam steering, laser energy attenuation and trapping of the incandescence signal [9].

One of the most uncertain areas of LII measurements in high pressures is the challenge of determining the boundary of the free molecular, transition and continuum regimes. These boundaries define the heat transfer model to be used for the interpretation of LII signals. Hofmann [10] reports that “the assumption of a free molecular regime is not valid for pressures above approximately 5 to 10 atm”. Therefore, the question for pressures higher than 10 atm is whether the regime is considered as a transitional or continuum regime. Thomson has considered pressures as high as 4.0 MPa to exist in the transition regime [9]. As for the results presented in this thesis, the McCoy and Cha transition regime heat conduction model [31] has been used for all pressures. This model has proved to result in 5% error of particle temperature at 80 atm [32].

Regardless of the used model, available literature suggest that at high pressures:
• LII intensities rise significantly due to the much higher soot concentrations produced in the flame,

• the conduction cooling rate of soot particles increases due to shifting to the transition regime by increasing the pressure,

• the soot morphology changes in a way which results in larger effective soot particle sizes with increasing pressure,

• the peak soot temperature decreases with increasing pressure primarily due to the particles’ enhanced heat conduction cooling rate and secondarily, due to the higher laser pulse energy attenuation due to higher soot loading in the flame [9].
Chapter 3

Theory

3.1 Soot Formation

Soot is formed during combustion processes in locally fuel-rich zones at elevated temperatures [17]. As an example, for laminar diffusion flames, soot inception zones occur within the wings of the flame where there is high fuel concentration [33]. These combustion generated particles, when mature, consist of small (15-50 nm diameter) carbon spheres which are held together by covalent bonds to form branched-chain aggregates of varied sizes [2] and contain up to 10% hydrogen on a molar basis [17].

The primary precursor to soot formation has been found to be polycyclic aromatic hydrocarbons (PAHs), which are cyclic molecules consisting of carbon atoms linked together with various hydrocarbon structures on the spokes of the wheel [34]. Like soot particles, PAHs are susceptible to heating by ultraviolet and visible light which could
cause them to incandesce and interfere with the signal from the soot particles of interest [35]. Since near-infrared light defeats the significant interference observed from PAHs and other hydrocarbons in the detection volume, 1064 nm lasers have become preferences for applications of LII [6, 7].

If PAH structures remain unoxidized, two possible pathways lead to their growth and transformation into soot particles: surface growth by addition of components from the gas phase and particle agglomeration caused by the coagulation of primary particles to large aggregates [17]. While surface growth leads to an increase in primary particle diameter (particles are approximated as spheres), coagulation leads to an increase in the size of the agglomerate [36]. Therefore, as soot particles rise through the flame, they increase in size up to a certain point where soot oxidation rates exceed formation rates. Once this point in the flame is reached, soot concentrations and particle sizes start to decrease [14]. As such, soot concentrations and particle sizes have a radial dependence that also changes with flame height.

### 3.2 Soot Properties

To process an LII signal, three properties of soot have to be known: density, thermal accommodation coefficient, and the absorption function of soot.

#### 3.2.1 Soot Density

One of the parameters involved in the calculation of soot particle size is its density. By approximating soot particles as graphite sheets, soot density, $\rho \text{ (kg/m}^3\text{)}$, is known to range from 1850 [37] to 2260 $\text{kg/m}^3$ [38]. Although temperature dependent densities may also be used in calculations, a constant value of 2030 $\text{kg/m}^3$ has been used in processing of LII signals throughout this thesis.
3.2.2 Thermal Accommodation Coefficient

The thermal accommodation coefficient, $\alpha$, is a measure of the energy transferred between a gas molecule and a surface during an interaction [39]. Larger values of $\alpha$ lead to a more efficient loss of energy to the surrounding atmosphere and, hence, a faster decay of the LII signal [6]. This property of soot is proportionally related to the primary particle size. Due to the vast uncertainty concerning the thermal accommodation coefficient, different models have used various values for this coefficient. These different values range from 0.2 to 1 [6]. In fact, many researchers evaluate this adjustable coefficient to match the measured primary particle size to physically-measured particles in identical flame conditions. It is worth mentioning that at high pressures, the regime of heat conduction shifts from free-molecular towards the continuum regime, thereby reducing the effect of the thermal accommodation coefficient [40].

3.2.3 Soot Absorption Function

$E(m)$ is a function of the complex index of refraction of soot, $m$. Although several studies have suggested that the soot absorption function might be wavelength dependent [16, 25, 41, 42], due to the vast uncertainties even in the sign of the slope of the wavelength dependence of $E(m)$, this function is assumed to be constant. The index of refraction is also suggested to be temperature-dependent [6]. Values of $E(m)$ commonly used by researchers range between 0.24 [6] and 0.4 [43] which are believed to be not affected by the pressure change.

3.3 Principles of LII

Essentially, LII involves the laser heating of an absorbing particle to a temperature below its sublimation temperature using a high-energy pulsed laser with the subsequent
photodetection of the incandescence resulting from the particle. This incandescence signal is the result of an intricate thermo-optical phenomenon, complicated by dependencies on particle size, number density of soot particles, particle temperature, surrounding ambient temperature, laser energy intensity, laser beam profile and other parameters [7, 44–46]. The basis of all models of LII is a transient energy balance on a single soot particle or aggregate. This energy balance describes the heat transfer between the particle and its surroundings, as well as the interaction of the particle with the incident laser radiation [7]. Whereas absorption of laser light heats up the particles, energy loss results from vaporization, heat conduction and radiation. Soot vaporization is the dominant heat-loss mechanism during the first $\sim$50 ns before heat conduction takes over. Hence, the time gate width considered for interpreting LII measurements usually starts after the first 50 ns have passed by to omit the evaporation term in the heat transfer equation. The radiative loss is usually negligible throughout the process [10, 32, 47]. It is the conduction term in the heat balance equation for soot particles which dominates the cooling process in LII techniques. It should be noted that the conductive cooling rate is proportional to the surface area of the particles which results in the greater contribution of large soot particles in the LII signal. In most models, aggregates of primary soot particles are considered as an individual primary particle with an equal mass. This simplified assumption is based on the premise that aggregates are groups of spheres touching at single points, thus keeping the surface area to volume ratio constant [43, 47–50]. For soot volume fraction measurements, the LII detected signal is temporally integrated over a few hundreds of nanoseconds after the laser pulse. Subsequently, time-resolved detection determines the decay rate of the signal which leads to the measurement of the size of the cooling soot particle.

The method used for deriving the soot particle size is based on the McCoy and Cha heat conduction model [31]. This model assumes that the velocity distribution of the gas molecules is independent of soot particle presence – that the Knudsen number is much
greater than one for the free molecular regime [31, 32]. Although increasing the pressure leads to moving from the transition regime towards the continuum regime, researchers such as Thomson et al. have used the same model for pressures up to 40 atm and have obtained consistent results with available literature [9]. For transition Knudsen numbers on the order of unity, another model, Fuchs’ method has also been recommended which considers particles as having two concentric layers. The inner layer is modeled as being in the free molecular regime while in the outer layer the heat conduction is modeled as existing in the continuum regime where collisions are frequent [32].

One of the most important factors in the magnitude of the LII signal is the laser fluence. This dependency is roughly linear up to the point where soot particles start to evaporate [47, 51]. At atmospheric pressure, this temperature is approximately 4000 K, which corresponds to about 0.3 to 0.4 $\text{J/cm}^2$ [6, 52]. Once this point is reached, increasing laser fluence leads to minor variations of the LII signal and a plateau region is observed on the excitation curve [10]. Generally, measurements are taken at whatever fluence results in the highest LII signal, which is usually the point where the soot particles just start to evaporate [13, 16, 27, 47, 52-54]. In the case of higher pressures, the vaporization temperature increases according to the Clausius-Clapeyron relation. Therefore, the laser fluence required for reaching equilibrium conditions will be higher than the fluence used for measurements at atmospheric pressure. Using a higher laser fluence is expected to result in higher LII signal intensities [10].

### 3.4 Analysis of Data

Between firing a laser at the target flame and obtaining meaningful measurements for the soot particle size and volume fraction, several steps should be taken. The first step involves the calibration of the photo multiplier tubes (PMTs) which detect the LII signal. Following this, gathered LII signals are processed to find temperature decay, particle size and soot volume fraction as functions of flame position.
3.4.1 Calibration

The PMTs have to be calibrated to provide the ability to produce meaningful data. This calibration helps to extract actual spectral radiance from the relative intensities resulting from the experiments. These extracted spectral radiances will then be compared to black body radiation curves to determine the temperature of the heated soot particles.

Previously, most LII researchers multiplied the LII intensity by a calibration factor to measure soot concentration in flames. This factor was usually obtained through correlating the LII intensity to a known soot volume fraction, which was either measured independently by light attenuation or gravimetric sampling, or simply known because a well-controlled carbon black generator or another known source was used [1]. Using this conventional method, obtained results should be relatively accurate as long as calibration conditions are close to those during LII measurements. However, any difference in flame temperatures, ambient pressure or flame conditions can lead to discrepancies. One of the most recently developed calibration methods has been presented by Snelling et al. [1] which eliminates the aforementioned limitations. Known as auto-compensating LII, this technique does not require any additional soot measurement tests to be conducted. Instead, a light source of know intensity is used to provide a source of illumination to the detection equipment of the LII setup. Based on this technique, for a given PMT gain voltage and incident light intensity, a unique voltage signal is produced by the PMT. By finding these three values, it is possible to obtain a calibration factor $\eta$ for each detection wavelength by

$$\eta = \frac{V_{\text{CAL}}}{R_S G_{\text{CAL}}}$$  \hspace{1cm} (3.1)$$

where $R_S$ is the spectral radiance of the light source and $V_{\text{CAL}}$ and $G_{\text{CAL}}$ are the associated PMT signal and gain voltages, respectively. In evaluating the temperature decay and soot volume fraction, this calibration factor is used.
Chapter 3. Theory

3.4.2 Temperature Decay

The first parameter that is calculated from the LII signal is the temperature of the soot particles. This time-dependent temperature is obtained after the laser is shot, where it is at its maximum, and shows a decreasing trend as time passes. Soot temperature is used in calculating the soot volume fraction and the decay rate is essential for determining soot particle diameter. The method used for finding the temperature of the cooling particles relies on two-colour optical pyrometry. In this method, it is assumed that all the soot particles in the laser probe are heated uniformly to the same temperature [1]. Using the calibration factor, the ratio of two signals of different wavelengths can be related to the ratio of power emission by the particles at each wavelength as

\[
\frac{P_p(\lambda_1)}{P_p(\lambda_2)} = \frac{\lambda_2^6 E(m_{\lambda_2})}{\lambda_1^6 E(m_{\lambda_1})} \exp \left[ -\frac{hc}{kT_s} \left( \frac{1}{\lambda_1} - \frac{1}{\lambda_2} \right) \right] = \frac{V_{\text{EXP}1} \eta_2 G_{\text{EXP}2}}{V_{\text{EXP}2} \eta_1 G_{\text{EXP}1}} \tag{3.2}
\]

where \(\lambda_1\) and \(\lambda_2\) are the two detection wavelengths which correspond to unique power emissions \(P_p\), index of refraction functions \(E(m)\), signal voltages \(V_{\text{EXP}}\), PMT gains \(G_{\text{EXP}}\), and calibration factors \(\eta\). The speed of light, Planck constant, and Boltzmann constant are represented by \(c\), \(h\), and \(k\) respectively, while \(T_s\) is the particle temperature. The index of refraction function is approximated as being constant between the two wavelengths. From Equation (3.2) the particle temperature can easily be found as an explicit function of the PMT signal ratio and thus a function of time:

\[
T_s = -\frac{hc}{k} \left[ \frac{1}{\lambda_1} - \frac{1}{\lambda_2} \right] \left[ \ln \left( \frac{V_{\text{EXP}1} \eta_2 \lambda_1^6}{V_{\text{EXP}2} \eta_1 \lambda_2^6} \right) \right]^{-1}. \tag{3.3}
\]
3.4.3 Soot Volume Fraction

As expected, higher soot loading in the flame leads to higher absorption of the laser power and an increased LII signal. In other words, a large signal corresponds to a large amount of radiating soot. Therefore, the volume fraction of soot in the probed volume is proportional to the magnitude of the signal produced by the PMTs at any given wavelength. Using the particle temperature and calibration factor from above, the soot volume fraction $f_v$ is calculated by

$$f_v = \frac{V_{\text{EXP}} \lambda_C^6 \exp \left( \frac{h\nu}{k\lambda_C T_p} \right) - 1}{\eta G_{\text{EXP}} 12\pi h c^2 E(m_\lambda) w_b}$$

where $\lambda_C$ is the centre wavelength of detection and $w_b$ is the width of the laser sheet [1].

3.4.4 Particle Size

Using the McCoy and Cha heat conduction model [31], the following expression for the effective primary particle size $d_p$ can be derived [9]:

$$d_p = \frac{12\kappa \gamma(T_g)}{\tau (d_p + G(T_g) \Lambda(T_g) C_s(T_s) \rho_s),}$$

where $\kappa \gamma(T_g)$ is the thermal conductivity of surrounding gas, $T_g$ and $T_s$ are, respectively, the local gas and effective soot temperatures, $\tau$ is the slope of $\ln(T_s - T_g)$ versus time.
over the considered time gate width after the peak LII intensity, $C_s(T_s)$ and $\rho_s$ are, respectively, the specific heat capacity and density of soot, and the heat transfer factor and mean free path of the combustion gas molecules are given, respectively, by

$$G = \frac{8f}{\alpha(\gamma + 1)}$$

and

$$\lambda_{\text{MFP}} = \frac{\kappa_a(T_g)}{\rho_s} \sqrt{\frac{\pi R_u T_g}{2W}}$$

where $\alpha$ is the thermal accommodation coefficient, $f = (9\gamma - 5)/4$ is the Eucken factor, $R_u$ is the universal gas constant, $W$ and $C_v(T_g)$ are, respectively, the mean molecular weight and the specific heat capacity of the surrounding combustion gas, $\gamma$ is the adiabatic constant, and $P$ is the pressure. Complete derivation of Equation (3.5) from the McCoy and Cha model has been presented by Cormier [22]. It is also shown that $d_p$ can alternatively be obtained using:

$$d_p = -\frac{1}{2}G(T_g)\lambda_{\text{MFP}}(P, T_g) + \sqrt{\left(\frac{G(T_g)\lambda_{\text{MFP}}(P, T_g)}{2}\right)^2 - \tau_T^{-1}\frac{12\kappa_a(T_g)}{C_s(T_s)\rho_s}}.$$  

At lower pressures, where $d_p \ll G\lambda_{\text{MFP}}$, an inexact explicit solution for $d_p$ can be found in which the approximate particle size does not depend on the thermal conductivity or adiabatic constant of the surrounding gas [22]:

$$d_p = -\tau_T^{-1}\frac{3\alpha P}{\rho_s} \frac{[2C_p(T_g) - R_u]}{C_s(T_s)} \sqrt{\frac{W}{2\pi R_u T_g}}$$

where $C_p$ is the heat capacity of the local combustion gas at constant pressure.
Chapter 4

Experimental Apparatus

4.1 General Layout

The apparatus consists of several optical and electrical components which can be categorized as excitation parts, high-pressure chamber and detection parts. Using the excitation components, the initial laser pulse is prepared to heat the particles in the flame and on the detection side, the subsequent LII signal is directed and detected. In addition to this, a shielding box is used to cover the detection parts which are prone to electromagnetic noise pickup. The high pressure LII optical configurations can be seen in Figure 4.1. This layout does not show electrical or support components including power supplies, oscilloscope, cables, and computers. A photograph of the apparatus (Figure 4.2) better illustrates the scale [22].
Figure 4.1: Schematic illustrating the optical layout of the two-colour LII apparatus setup for high pressure measurements. Oscilloscope, power supplies, cables, and gas delivery system are not shown.
Figure 4.2: A photograph showing the optical components of the LII apparatus set up for high pressure measurements. Detection optics are on the foreground table and excitation optics including partial laser head are visible on the background table. Oscilloscope, power supplies, and fuel and gas delivery system are not in full view. In the background, the high pressure chamber is partially visible between the excitation and detection optical tables. The electromagnetic shielding box that is normally present is omitted so that optical components are visible. Metal shielding boxes built around the detectors are, however, present [22].

4.2 Support and Mounting

The apparatus’s optical components are mounted on several optical breadboards which are secured to mobile aluminum tables. The design purpose for this support system was to provide fixed and precise positioning of optical components all the while preventing the potential long-term vertical deflection of the table frames.
The table frames supporting the optical breadboards consist of 60 mm cross-section aluminum framing manufactured by Bosch Rexroth. The thick cross section enables the frames to support several hundred kilograms of equipment. The tables ride on rubber caster wheels to provide mobility between experiments. Once the tables are manually lifted high enough, the weight is transferred on to the metal feet and the position is fixed. The feet are independently height-adjustable to allow the optical breadboards to be level during experiments. Unlike the atmospheric setup, for high pressure measurements, one table with two breadboards is used for the detection optics and a second table with a single breadboard is used for the excitation optics.

Optical components are mounted on several Thorlabs PerformancePlus™ series optical breadboards.

4.3 Excitation

The excitation part of the apparatus includes the laser and the beam-manipulation optics which adjust the laser power and shape and focus the beam on the desired section of the flame.

4.3.1 Laser

Although working with an invisible infrared laser beam adds to complications, several groups have found that significant laser-induced florescence is induced at 532 nm [6, 35]. Due to this interference caused by beam absorption of PAHs in the visible and
ultraviolet, an Nd:YAG laser operating at 1064 nm is used in the apparatus. The laser is a multimode Continuum Surelite II-10 modified to use a graded reflectivity mirror (GRM). This modification allows for the superpositioning of different modes which results in a stable super-Gaussian spatial profile that has a flatter centre peak than a normal Gaussian distribution.

The laser has a measured divergence of $5 \times 10^{-4}$ radians and a 3.6 mm initial $1/e^2$ spot size. With a 0.18 m cavity length and 1 cm$^{-1}$ laser linewidth, the Rayleigh range\(^1\) of the laser beam comes to 3.83 m.

The laser should at all times be connected to a 240 V, 15 A power source. As such, the laser’s main breaker switch should be kept closed so that the system has continuous power. Instructions on operating the laser can be found in the manufacturer’s operation and maintenance manual [55, p. 25].

### 4.3.2 Half Wave Plate

To ensure maximum heating while preventing soot sublimation, a maximum fluence of 0.3 J cm$^{-2}$ has been suggested [6]. Therefore, the default laser output, over 2 J cm$^{-2}$, is more powerful than is needed. To attenuate the beam power, without altering the spatial and temporal profiles of the laser beam, a half wave plate (HWP) is used to reduce the laser output. The Thorlabs half wave plate can be rotated to alter the portion of p-polarized light that is rotated to s-polarized light. Anywhere between all of and none of the p-polarized light will be rotated depending on the HWP rotation angle. The laser beam’s fluence as a function of the half wave plate’s angle of rotation is given by:

\[^1\]The propagation distance from a beam’s waist at which the cross sectional area of the beam has doubled.
\[
\frac{\Phi}{\Phi_0} = \frac{1 + \sin 4\psi}{2}
\] (4.1)

where \(\Phi / \Phi_0\) is the fraction of the maximum beam fluence \(\Phi_0\) that is transmitted as a function of the half wave plate rotation angle \(\psi\) in radians. A graph illustrating the relation between the HWP rotation angle and the resulting laser power has been provided by Kempthorne [21]. For the set of experiments presented, the HWP rotation angle was set to 169°. Note that the half wave plate should be angled approximately 5° from perpendicular to the incoming laser beam to prevent any potential reflection of the laser beam which could damage the laser cavity.

4.3.3 Thin Film Polarizer

Once the half wave plate rotates a portion of the p-polarization to s-polarization, the Thorlabs thin film polarizer, reflects all s-polarized light into a nearby beam dump and allows the remaining light to pass through without an alteration in beam profile. To work properly, it is essential that the thin film polarizer be rotated to Brewster’s angle\(^2\) from the beam’s propagation axis. By changing the portion of s-polarized light transmitted through the half wave plate, the portion of light transmitted through the thin film polarizer is changed, thereby the combination of the half wave plate and the thin film polarizer leads to attenuation of the laser beam.

4.3.4 Aperture

The detection volume is primarily defined by the excitation beam shape. To shape the beam, a small slit is used which is referred to as the excitation aperture. The

---

\(^2\)The angle of incidence for a transparent dielectric surface at which light will be either perfectly transmitted or reflected depending on if the light has a particular polarization or not, respectively. Roughly 56° for glass in air.
size and shape of this aperture determines the cross section of the plane in the flame that will be irradiated. For high pressure measurements, a highly light-tolerant alumina ceramic 3 mm × 100 µm (± 10 %), 125 µm thick slit manufactured by Lenox Laser was used to define the shape of the laser beam to be imaged onto the detection volume within the flame. Even though ceramic apertures are recommended due to their high damage threshold, prolonged bombardment with high intensity light can result in damage to the slot, leading to its malfunction. Therefore, in addition to controlling the laser fluence using the half wave plate and thin film polarizer, inspection of the slit, prior to experiments, is recommended.

By using a beam profiler, the diffraction caused by the thin slit is detectable at the burner location; however, the intensities of non-primary maxima are small (< 5 % total fluence).

4.3.5 Relay Lens

To image the slit of light into the flame, a relay lens is used. This lens focuses on the intended measurement point within the flame while creating an inverted image of the flame on the excitation aperture.

For high pressure measurements, a f/16 400 mm focal length achromatic lens with an anti-reflection coating for infrared wavelengths is used to produce unity magnification of the flame. Although the large size of the high pressure chamber dictates longer focal lengths, higher f-numbers lead to less detected light. The calculated depth of field of the 400 mm lens is roughly 0.63 mm. To provide an almost clear image of the flame on the aperture, the lens has to be placed between the aperture and flame with 40 cm spacings.
4.4 Combustion Chamber

A high pressure combustion chamber was used to study the effect of pressure on soot formation. The burner is similar to one used by Miller and Maahs [56] for its high flame stability. The chamber has a maximum design pressure of 110 atm. Complete technical drawings of the chamber can be found in the thesis of Mandatori [57]. The chamber is closed at both ends and optical access into the chamber is provided through three quartz glass viewports located at $0^\circ$, $90^\circ$ and $180^\circ$. The two viewports used for LII applications are located $90^\circ$ apart and the third is used for flame observation at times when the laser power is off. To prevent potential damage caused by the laser beam passing through the flame and escaping the chamber, it is suggested that burn paper is placed in front of the third viewport while the laser power is on.

The chamber is mounted on a three-axis translational system that moves with respect to the optical diagnostic system. Each translation stage has a positional precision better than 0.005 mm. The gas delivery system is as previously described by Joo for methane/air flames [58, pp. 19-29].

4.5 Detection

The LII signal emitted from the heated soot particles has to be collected, manipulated and recorded at two distinct colors. Therefore, optical components focus the produced signal, split it into two different wavelengths and transmit the resulting beam to two detectors. Each of these detectors measure a color-filtered portion of the total emitted light spectrum. This light is then transferred to voltages by the detectors, and measured by an oscilloscope to produce the LII signals.
4.5.1 Relay Lens

This lens, which is usually paired with an almost identical one on the excitation side, is manufactured by Thorlabs. The imaging of the detection volume occurs in the same way that the laser light is relayed to the flame. This relay lens images the desired detection volume on to an aperture on the detection side, namely the detection aperture. Once the position of the lens is correctly adjusted between the detection aperture and the flame according to the focal length (which in this case is 400 mm), an upside down image of the flame can be observed on the detection aperture.

Generally, relay imaging is susceptible to creating a slightly softer (or more out-of-focus) detection volume since both the excitation and detection relay lenses must be manually focused to a single vertical axis in space. While larger f-numbers increase the lens’s depth of field resulting in less alignment sensitivity and greater measurement accuracy (since the detection volume will be in sharper focus for all measurement positions), the signal to noise ratio is dependent on the distance. In other words, larger f-numbers result in longer distances required for collecting signals which leads to reduced collected signals and thus lower measurement sensitivity. Thus, the minimum soot volume fraction detectable by lenses with high f-numbers, such as the ones used in the high pressure setup, is higher than in conditions where application of lenses with lower f-numbers are feasible.

The detection-side relay lens is identical to the excitation-side with the exception of having visible-wavelength anti-reflective coating rather than infrared.

4.5.2 Aperture

By focusing the image of an aperture onto the laser-heated particles within the flame, the detection volume is ultimately defined. The shape of the detection volume is dictated
by the shape of both the excitation and detection apertures. In other words, the volume in which the soot formation parameters are measured is defined as the intersecting shape of the image formed on the excitation aperture and the image formed on the detection aperture.

A Lenox stainless steel aperture is used to image the detection volume with respect to the detection optics. For high pressure measurements, a vertical $3 \text{ mm} \times 50 \mu\text{m} (\pm 5\%)$ slit was used; the intersection of the images of the excitation and detection apertures define a $50 \mu\text{m} \times 50 \mu\text{m} \times 3 \text{ mm}$ vertical line as a $7.5 \times 10^{-12} \text{ m}^3$ detection volume. Although a detection volume of this size is convenient for some flames, this is not the case for others. A methane/air laminar flame with a volumetric fuel flow rate of $50 \text{ cm}^3/\text{min}$ results in a flame height of $9 \text{ mm}$ for pressures between 10 and 100 atm [58]. Thus, a $3 \text{ mm}$ tall aperture images one third of the flame as the detection volume. Due to the large amount of variances which exist in one third of a flame, a modification to the aperture was necessary. Generally, a smaller detection volume leads to more diffraction and less signal but higher spatial resolution. Therefore, to reach an optimized tradeoff between the signal to noise ratio and spatial resolution, $2/3$ of the detection aperture was blocked in such a way that a detection volume of $50 \mu\text{m} \times 50 \mu\text{m} \times 1 \text{ mm}$ vertical line was achieved. The smaller detection volume at high pressures means higher precision and smaller signals. Because the high pressure flames are smaller and contain more soot than atmospheric flames, this is an apt compromise.

### 4.5.3 Dichroic Filter

Since the soot particles’ temperature is obtained through two-color pyrometry, which requires two distinct signals with different wavelengths, the emitted light must be split
into two beams. After an achromatic lens is used to collimate the broadband LII signal imaged from the detection aperture, the light is passed through a Thorlabs short-pass dichroic mirror that transmits light below 488 nm and reflects the remaining incident beam.

4.5.4 Band Pass Filters

Since each beam must be filtered for a single color, the split signal then passes through two Thorlabs 40 nm full width at half maximum (FWHM) band-pass filters centered at 440 and 692 nm, respectively. Each beam is then focused to a photomultiplier tube (PMT) by another achromatic lens resulting in unity magnification. These wavelengths were chosen to avoid interference of C\textsubscript{2} Swan band emissions at 473 nm, 516 nm, 563 nm, and 618 nm [6, 59, 60]. Although shorter wavelengths are advantageous to allow easy discrimination against flame radiation, the large separation between the detection wavelengths allows for more accuracy of the two-color pyrometry technique [6].

4.5.5 Photomultiplier Tubes and Circuit Boards

Due to the short lifetime of the LII signal and the importance of time-resolved measurements in LII, fast photo-detectors are required. Charge-coupled devices (CCD) lack the required temporal resolution, and so photomultiplier tubes are used.

The Hamamatsu PMTs have peak detection efficiency between 400 and 800 nm which fits the 440 nm and 692 nm detection colours. The PMTs have a rise time of 1.4 ns which accommodates the Nyquist-Shannon theorem which implies that sampling should occur
at a frequency of half the electronic rise time. The PMTs are matched to the designated wavelengths and mounted on circuit boards manufactured by Artium, a company specializing in the production of commercial emissions diagnostics LII apparatuses. These boards feature noise filters and op-amps that help to reduce signal noise from the PMTs. In addition to this, the PMTs and circuit boards are completely enclosed within a 6.4 mm thick conductive box to eliminate any noise produced by the detection electronics themselves.

4.5.6 Power Supply

Each PMT requires a mainline ±15 V supply in addition to a variable gain between 0 and 5 V. The gain voltage is manually set before taking measurements, and is the same for both PMTs. The circuit board electronics within the PMT boxes boost this voltage by a factor of 200 to provide the high voltage required by the PMTs. A single GW Instek GPS-4303 quad-output power supply is used for supplying the power for the PMTs. This power supply is kept within the same shielding box as the detection optics to shield exposed wire leads. The correct channel configurations are as in Table 4.1. As it can be seen, channel 4 and channel 1 of power supply provide the required −15 and +15 V required, respectively, while the gain voltage is controlled manually through the positive port of channel 2. Positive port of channel 4 and negative ports of channel 2 and channel 1 are all grounded using the wire connected to the ground port of the oscilloscope.

4.5.7 Oscilloscope

To measure the fast signals being produced by the PMTs, a highly responsive oscilloscope is needed to illustrate, store and analyze the signal. The LeCroy Wavesurfer
Chapter 4. Experimental Apparatus

Table 4.1: Power supply channel configuration for each of the PMTs’ input wires.

<table>
<thead>
<tr>
<th>Insulation Colour</th>
<th>Plug Colour</th>
<th>Role</th>
<th>Power Supply Channel</th>
</tr>
</thead>
<tbody>
<tr>
<td>White</td>
<td>White</td>
<td>+15 V</td>
<td>Both to Channel 1 Pos</td>
</tr>
<tr>
<td>Red</td>
<td>Red</td>
<td>-15 V</td>
<td>Both to Channel 4 Neg</td>
</tr>
<tr>
<td>Black</td>
<td>Black</td>
<td>Gain</td>
<td>Both to Channel 2 Pos</td>
</tr>
<tr>
<td>Black</td>
<td>Brown</td>
<td>Ground</td>
<td>Both to Ground</td>
</tr>
<tr>
<td>Bare</td>
<td>Black</td>
<td>Shielding</td>
<td>Both to Ground</td>
</tr>
</tbody>
</table>

64Xs oscilloscope, which is used in the current setup, has a 600 MHz bandwidth, sampling rate of $2.5 \times 10^9$ samples per second, and 11 bits of vertical resolution. The oscilloscope monitors both PMT outputs in addition to their gain voltage. Shielded bayonet Neill-Concelman connector coaxial cables are used with the oscilloscope and have a 0.75 velocity factor, yielding approximately 4 ns of temporal offset between signals per meter of path difference. This offset is insignificant because cable lengths are equal to within less than a centimeter. Generally, channel 1 and channel 2 port of the oscilloscope correspond to the 440 nm and 692 nm PMTs, respectively.

4.5.8 Shielding Box

Shielding detection equipment from electromagnetic interference (EMI) is crucial; visible light leaks will artificially inflate the LII signal and decrease the signal-to-noise ratio of measurements. In addition to visible light, invisible EMI is of greatest concern; interference can be caused by any electronic equipment in the room including lights, motors, power supplies, and the LII laser.

Since experiments revealed major EMI, a shielding box made of steel sheet with thickness of 1.5 mm was designed and manufactured, as recommended by Cormier [22],
to absorb and reflect low frequency electromagnetic waves. The detection optics along with the PMT boxes and power supply are placed within the shielding box where a sheet separates the power supply from the other detection equipments. Additionally, the box was designed in such way as to cover the oscilloscope’s ports from potential electromagnetic noises.
Chapter 5

Experimental Procedure

5.1 Overview

Taking measurements using the laser-induced incandescence (LII) apparatus and obtaining meaningful data can be divided into six primary categories: Optical Calibration, Setup, Alignment, Fuel Flow Calibration, Taking Measurements, and Data Analysis.

5.2 Optical Calibration

To extract actual spectral radiance from the relative intensities obtained from the PMTs, an optical calibration is required. This calibration will yield a constant parameter associated with each detector that will be used to calculate soot volume fraction and
particle temperature (which is then used for particle size measurements). The detectors are the only part of the apparatus that require calibration. In fact, calibration must be performed whenever any of the detection-side optical components are altered or moved in the slightest.

One of the most recently developed calibration methods presented by Snelling et al. has been used to obtain the calibration factor. One of the advantages to this calibration technique [1] is that it accounts for the many defects that could otherwise be hindering the detection optics. In other words, as long as a defect is consistent across all measurements and does not show bias to a particular portion of the detection volume, the calibration will often negate the problem.

### 5.2.1 Components and Setup

In addition to the LII detection equipment, the calibration apparatus consists of very few components: an integrating sphere (an incandescent light source of uniform and high intensity), a power supply, and a spectrometer with software to quantify the light source’s emissions.

**Integrating Sphere**

The integrating sphere used is a halogen lamp-illuminated SphereOptics SPH-6-2 (serial number 3925). It has a 15.2 cm inner diameter spherical Optowhite\textsuperscript{TM} Lambertian surface\textsuperscript{1} with two ports. In addition to a 1 mm diameter fibre optic port used for

\textsuperscript{1}A surface that adheres to Lambert’s cosine law; its observed scattered light intensity is independent of the angle of observation.
spectrometric measurements, it has a 3.81 cm diameter knife-edge port which is used to illuminate the LII detectors during calibration.

The integrating sphere will output a predictable and steady spectrum of light. The spectrum is not truly blackbody but is meant to simulate an LII signal. The light simulates the voltage responses seen by the PMTs at two particular wavelengths.

Ideally, the integrating sphere’s output port should be in the same position as the flame to be measured. Actual placement of the integrating sphere depends on the burner being used. Since the integrating sphere is housed in a large, unwieldy cube, placing it inside the high pressure combustion chamber is impossible. Therefore, it is suggested to calibrate before positioning the detection optics for LII measurements. The sphere can then be placed in front of the detection optics at a distance equal to that during measurements. A spare quartz viewport has to be placed in front of the detection optics in the line of sight of the integrating sphere. This will simulate the chamber’s viewport’s presence in actual measurements conditions. Preferably, the spare window will be placed in the same relative position as would be the case during LII measurements.

**Power Supply**

An Agilent E3634A 200 W 0-25 V, 7 A / 0-50 V, 4 A DC power supply was used to supply the integrating sphere lamp with a steady current of 4.166 A at 25 V. This power supply should be properly grounded and connected to the integrating sphere via the sphere’s two gator clips.

**Spectrometer Software**

The spectrometer collects and measures light directly from the integrating sphere during calibration. Measurements are sent to a computer, averaged over time, and plotted using the SMS-500 software.
A 1 mm fibre optic cable is connected to both the integrating sphere and spectrometer to collect light, and the spectrometer is plugged into a computer with USB port and proprietary SphereOptics SMS-500 spectrometer software installed. For LII purposes, the oscilloscope is used as the computer and the software is loaded onto it. This software produces a text file containing a table of precise time-averaged spectral radiance values as a function of emission wavelength. The spectral radiance values corresponding to the detected center wavelengths are used to determine the calibration factor for LII analysis.

5.2.2 Experimental Procedure

The calibration procedure is straightforward and requires a small set of measurements. Two distinct data sets are acquired: the integrating sphere output spectrum, and the PMT voltage response curve.

Integrating Sphere Output Spectrum

Once the integrating sphere has been connected to the power supply, it has to be turned on. Quickly switching the integrating sphere lamp on at full power can cause damage to the filament. Therefore, the power supply should be slowly dialed up from zero to the required amperage (roughly 4.166 A) until the potential sits at 25 V. This process should be done over a period of one to two minutes. This process should be repeated in reverse when powering down the lamp after calibration.

Before the integrating sphere spectrum is obtained, the spectrometer must be calibrated. This is a simple procedure done by the spectrometer’s software. To do this,
first connect the spectrometer to the oscilloscope using the USB cable and open the installed SMS-500 software. Then, make sure the spectrometer input cap is closed, so that it sees no light. With the software, select Tools $\rightarrow$ System Zero to calibrate dark current. This takes several minutes. Once finished, connect the integrating sphere to the spectrometer using the fibre optic cable.

To configure the number of spectrum samples, select Setup $\rightarrow$ Setup Spectral Parameters. The settings found in Table 5.1 are sufficient.

Table 5.1: Spectrometer parameters used during calibration.

<table>
<thead>
<tr>
<th>Setting</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Integration Time</td>
<td>2 ms</td>
</tr>
<tr>
<td>Samples</td>
<td>5 - 15</td>
</tr>
<tr>
<td>Boxcar Smoothing</td>
<td>2</td>
</tr>
<tr>
<td>Auto Range</td>
<td>On</td>
</tr>
<tr>
<td>Wavelength</td>
<td>335 - 1100 nm</td>
</tr>
</tbody>
</table>

To preview a spectrum scan, select Start Auto Scan, wait a few moments, and then select Stop Auto Scan. A spectrum should be shown. To obtain the full spectrum measurement, select Acquire Data. You will then have to enter an operator’s name and confirm. To save the data set to a text file (in the software’s installation folder), select Save New Data. To save the spectral data to a desired location, once the auto scan has been stopped, select File $\rightarrow$ Save Spectral Scan. The file location can then be determined and the file is saved with an .scn file extension.

PMT Voltage Response Curve

The spectrometer can be put away and disconnected from the integrating sphere at this point. Once the detection optics, the integrating sphere and the viewport are
correctly positioned, the PMTs have to be connected to the detection power supply and the oscilloscope via corresponding cables. Before the power supply’s output switch is turned on, make sure the gain voltage supplied to the PMTs is lower than 3 V. Exposing the PMTs to higher gain voltages can result in irreversible damage to the circuit boards.

The gain voltage should initially be set low, near 2.3 V as measured by the oscilloscope and increased by roughly 0.1 V for each new data point. It should be noted that gain voltages shown by the power supply are not accurate. Therefore, the accurate gain voltage at each point should be measured using the oscilloscope. At each gain voltage, the corresponding voltage response and dark current voltage for each PMT should be recorded. The dark current can be obtained by completely blocking the entrance to the detection optics’ shielding box. Collecting points up to between 2.7 and 3.0 V gain is sufficient, yielding between 5 and 10 data points.

5.2.3 Using the Data

The calibration factor at any given gain voltage is given by

\[ \eta = \frac{V_{\text{CAL}}}{R_S G_{\text{CAL}}} \]  \hspace{1cm} (5.1)

where \( R_S \) is the spectral radiance of the light source and \( V_{\text{CAL}} \) and \( G_{\text{CAL}} \) are the associated PMT signal and gain voltages, respectively. Therefore, for a given PMT gain voltage and incident light intensity, a unique voltage signal is produced by the PMT. The first step is to obtain the PMT voltage response curve. The PMT response voltage \( V_{\text{CAL}} \) is calculated
Chapter 5. Experimental Procedure

by subtracting the dark current voltage at \( G_{\text{CAL}} \) by the measured response voltage. The response voltages can then be plotted against the corresponding gain voltages. The slope of this plot should be positive and linear and can be used to interpolate the response voltage for any given gain voltage. For any new gain voltage used in LII experiments, the corresponding changes should be made accordingly to the LII software used for analyzing the data.

The next step to determine the calibration factor is to obtain the spectral radiance. The spectral radiance is listed in the table output by the spectrometer software. When reading the spectral radiance from the output file, the correct value is not given by the detection wavelength but an average across the range of wavelengths. This is because the filters allow light to pass as a function of wavelength across a narrow spectrum. For example, the 440 nm filters used in the apparatus have a true center wavelength of 443 nm based on the transmission spectrum supplied by the manufacturer and transmit significant amounts of light between 415 and 464 nm. As such, the 692 nm filters have a true center wavelength of 690 nm. Generally, to obtain these values, the net transmission spectrum should be multiplied by the corresponding integrating sphere spectral radiances; the resulting spectral radiance should be averaged over the sum of wavelength transmittances to obtain a single value for that color. The spectral radiances at 443 and 690 nm can then be integrated into the LII software.

Thus, in summary, from the gain voltage used during the LII measurements, the response voltage for each detection channel is interpolated and the spectral radiances corresponding to the two center wavelengths are read. By integrating these 4 values into the LII software, the data can be analyzed.
5.3 Setup

The LII apparatus setup is explained in the previous chapter. Nonetheless, several settings of the oscilloscope have to be examined before detection of LII signals.

5.3.1 Channel Vertical Adjust

The Channel Vertical Adjust window contains the setting options for signal interpretation. Appropriate settings for measurements are as in Table 5.2. **Averaging** should typically be set to 1 unless taking final measurements. For final measurements, averaging of 100-400 shots is recommended.

<table>
<thead>
<tr>
<th>Setting</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Volts/div</td>
<td>Adjust to fit signal</td>
</tr>
<tr>
<td>Variable gain</td>
<td>No</td>
</tr>
<tr>
<td>Offset</td>
<td>Adjust to fit signal</td>
</tr>
<tr>
<td>Bandwidth</td>
<td>Full</td>
</tr>
<tr>
<td>Invert</td>
<td>No</td>
</tr>
<tr>
<td>Coupling</td>
<td>DC50Ω</td>
</tr>
<tr>
<td>Deskew</td>
<td>0</td>
</tr>
<tr>
<td>Probe Attenuation</td>
<td>÷1</td>
</tr>
<tr>
<td>Averaging</td>
<td>100 to 400 sweeps</td>
</tr>
<tr>
<td>Interpolation</td>
<td>Linear</td>
</tr>
<tr>
<td>Noise Filter</td>
<td>None</td>
</tr>
</tbody>
</table>
5.3.2 Timebase

Suitable time resolution (Time/Division) for LII signals is 100 ns with a delay of approximately -350 ns.

5.3.3 Trigger

Detecting the LII signal is done automatically by the oscilloscope if the trigger is setup correctly. For measurements, the triggering mode should be changed from Auto to Normal. This allows for measurements to take place only if the trigger value is reached. Correct setup of the trigger value requires tuning between laser shots. Once the correct value has been reached, the LII signal will be detected and shown on the oscilloscope monitor as a peak in the light detection. If a signal is found, the laser should be set to automatically fire at 10 Hz until the oscilloscope collects enough sweeps for averaging.

Suitable trigger settings are as in Table 5.3. Source should be the larger of the two channels, which is typically C2 (692 nm).

Table 5.3: Oscilloscope trigger settings used for LII measurements.

<table>
<thead>
<tr>
<th>Setting</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Type</td>
<td>Edge</td>
</tr>
<tr>
<td>Source</td>
<td>C2</td>
</tr>
<tr>
<td>Level</td>
<td>Half of peak signal</td>
</tr>
<tr>
<td>Slope</td>
<td>Positive</td>
</tr>
<tr>
<td>Coupling</td>
<td>DC</td>
</tr>
</tbody>
</table>
5.4 Alignment

Alignment procedures have been presented by Cormier [22].

5.5 Fuel Flow Calibration

Before setting up the flame, the fuel flow rate has to be calibrated to ensure accuracy. Since the fuel flow rate has a dominant effect on the visible flame height, this step is vital in obtaining a flame with the desired properties. As for measurements on methane flames with a fuel flow rate of 50 cm$^3$/min, a visible flame height (indicated by soot radiation emission) of about 9 mm has to be achieved for pressures between 10 and 100 atm [58].

5.6 Taking Measurements

Once the flame has been set up, the chamber has to be pressurized to reach the desired pressure. During this time, it is suggested for the laser to be powered on (with shutter closed) to allow warm up time for the laser. Without this warmup, the shutter switch does not function.

When the chamber reaches the desired pressure, the Y and Z axes of the burner’s movement stage has to be adjusted in a way that the centerline of the top 3 mm of the flame coincides with the excitation aperture. With correct alignment and by adjusting the X axis, the same can be done with the detection aperture. The only difference is that
the detection aperture only transmits the top 1 mm of the flame image since the upper 2/3 of the aperture is blocked.

Before shooting the laser and detecting signals, the gain voltage supplied to the PMTs has to be fixed. Higher pressure results in higher soot loading and thus, larger signals. If the gain voltage is set too high for these pressures, the PMTs will reach DC saturation (DC saturation can be detected when the signals shown by the oscilloscope have a flat peak compared to the normal sharp peak) that can cause damage to the circuit boards. Therefore, a lower gain voltage should be used for higher pressures. For methane flames at 10, 20 and 40 atm, gain voltages around 3, 2.8 and 2.4 V, respectively, are recommended. At this point, by shooting the laser and observing the resulting peak in light detection from the oscilloscope (namely, the LII signal), it can be confirmed that components of the apparatus are functioning correctly.

Since measurements are usually done at a certain height along the flame, it is necessary to determine the Y coordinate of the tip of the flame. If the laser is set to pulse mode, by moving the burner’s movement stage at the same time, the tip of the flame can be determined. Once the LII signal is lost, the tip of the flame is reached. For the aforementioned methane flame, 6 mm height above burner (HAB) corresponds to 3 mm into the flame with reference to the tip of the flame. To ensure consistency, this step can be repeated for different pressures; if the tip of the flame’s Y coordinate changes, the flame height has been altered and the chamber should be examined for potential fuel line leaks.

Measurements are typically taken along the flame axis or perpendicular to it to obtain centreline or profile measurements, respectively. For profile measurements, the initial position can be on either edge of the flame at the desired HAB. The coordinate of the
edge of the flame can be obtained through the same procedure used for the tip. Although the burner stage can be moved in both the X (axis of laser beam) and Z (axis of detection optics) direction between measurement positions, profile measurements should be done in the same plane as the excitation laser sheet. This means moving the burner stage in the X axis for different measurement positions. This is to sacrifice laser attenuation for a lower average signal attenuation.

Step sizes for measurement positions are typically equal to the height or width of the detection optics’ aperture. Step size can be smaller than the aperture if the aperture is large, yielding a rolling average. The measurements are, therefore, done in 50 μm intervals.

It should be noted that for all measurements, it is essential for the signal to fit on the oscilloscope’s screen. If the resulting signal is too large, by changing the vertical setting from mV to V, the vertical scale can be adjusted accordingly.

5.6.1 Saving Data

At each measurement position, after the oscilloscope has averaged a suitable number of sweeps, the signals may be saved (File → Save Waveform...). The settings are found in Table 5.4. For each measurement, two files must be manually saved: one for Source set to C1 and another for C2. The file, position, and directory formats are dictated by the analysis software.

5.7 Data Analysis

The first step to analyzing the data is adjusting the calibration information based on the gain voltages used in the available program. Then, the data analysis can be performed
Table 5.4: Oscilloscope file settings used for LII measurements.

<table>
<thead>
<tr>
<th>Setting</th>
<th>Value or Format</th>
</tr>
</thead>
<tbody>
<tr>
<td>Save To</td>
<td>File</td>
</tr>
<tr>
<td>Source</td>
<td>C1 or C2</td>
</tr>
<tr>
<td>Trace Title</td>
<td>PosN&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
<tr>
<td>Format</td>
<td>Excel</td>
</tr>
<tr>
<td>SubFormat</td>
<td>Time &amp; Amplitude</td>
</tr>
<tr>
<td>Auto Save</td>
<td>Off</td>
</tr>
<tr>
<td>Save files in directory</td>
<td>.../MonDDYYYY/Raw</td>
</tr>
</tbody>
</table>

<sup>a</sup> Pos1, Pos2, …, PosN for N measurement positions

automatically by the analysis software. The software will output time-resolved temperature, soot volume fraction, and particulate size excel spreadsheets for each measurement position as well as position-resolved time-averaged spreadsheets for plotting.

The software, implemented with Python, requests experiment-specific parameters upon the first attempt to analyze a data set. These parameters are saved in a CSV file in the data set’s folder for future reference. If any of these parameters need to be changed, this file is adjusted by using the Modify Data Set Information option and performing a full analysis of the new data set. Physical constants that do not change from experiment to experiment are saved in the program script itself, and can be easily edited. Calculations for temperature, soot volume fraction, and particle size are done automatically in series. Temperature, soot volume fraction, and particle size are calculated as functions of time for each location and output as separate CSV files for each location. A single number for each soot volume fraction and particle size is then averaged for
each measurement position; these are collated and saved in a soot volume fraction CSV file and particle size CSV file. More detailed information about each of the numerous functions in the software can be found in the software’s inline documentation.

During analysis, it may prove necessary to analyze data sets with different values for the thermal accommodation coefficient, soot absorption function and the start and end time considered after the laser shot, to achieve consistent results. Generally, time windows closer to the peak signal are preferred due to the high signal to noise ratios at these times. However, moving away from the laser peak results in minimizing effects of sublimation, which are not considered in the thermal model used for measurements [47]. Therefore, an optimum time gate width for the soot volume fraction and particle size measurements can be found accordingly.

Parameters used in the analysis of the signals are as in Table 5.5.

5.8 Maintenance

The apparatus is largely maintenance free; the primary exception is the laser itself. In addition to laser maintenance, the optical components must be inspected and cleaned before each experiment. Covering all optics after experiments is necessary to prevent dust collection. In case of dust presence on optics, cleaning is done by wetting a new lens cleaning tissue with isopropyl alcohol and gently dragging the tissue along the surface of the glass. To avoid scratching the surface, do not re-use the tissue and do not apply pressure while cleaning. To avoid leaving residue or damaging optical coatings, only isopropyl alcohol should be used.
Table 5.5: Parameters used for high pressure methane/air LII analysis.

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E(m)$</td>
<td>0.4 [^{[43]}]</td>
</tr>
<tr>
<td>$\alpha$</td>
<td>0.26 [^{[9]}]</td>
</tr>
<tr>
<td>$W$</td>
<td>0.02896 $\text{kg mol}^{-1}$</td>
</tr>
<tr>
<td>$T_g$</td>
<td>1700 K [^{[58]}]</td>
</tr>
<tr>
<td>$\rho_s$</td>
<td>2030 $\text{kg m}^{-3}$ [^{[61]}]</td>
</tr>
<tr>
<td>$\kappa_a$</td>
<td>Temperature-dependent [^{[62]}]</td>
</tr>
<tr>
<td>$C_s$</td>
<td>Temperature-dependent [^{[63]}]</td>
</tr>
<tr>
<td>$C_p$</td>
<td>Temperature-dependent [^{[64]}]</td>
</tr>
<tr>
<td>Time window</td>
<td>20 – 80 ns for SVF</td>
</tr>
<tr>
<td></td>
<td>70 – 100 ns for particle size</td>
</tr>
</tbody>
</table>

\[^{[a]}\] Value for atmospheric air

\[^{[b]}\] Value for graphite

The laser maintenance is straightforward and covered in detail within its user manual. The most frequent maintenance is ensuring that the laser’s breaker switch is always closed so that there is power to the system. The water level should also be checked every few months and the flash lamp replaced as required. For detailed procedures and maintenance schedules refer to the laser’s manual \[^{[55, p. 63]}\].
Chapter 6

Results

6.1 Signal Quality

For all pressures considered (10, 20 and 40 atm), the signal quality for both wavelengths showed satisfactory results up to 100 ns after the peak LII signal. After 70 ns from the peak signal, the signal to noise ratio visibly decreased, thereby preventing smooth decays. An example of the disturbances appearing in the signals can be seen in Figure 6.1 for a signal obtained at 20 atm. Although the relative magnitudes of disturbances were small, significant deviations from actual values of soot temperature, soot volume fraction and particle size can result from these noises.

6.2 Temperature

To prevent soot sublimation, temperatures resulting from laser heating should stay below 4000 K [47]. Formerly, soot temperatures as high as 4600 K were obtained. By
changing the half wave plate settings, the laser power was reduced, resulting in lower peak temperatures. Figure 6.2 shows the time-resolved temperatures calculated using the raw signal shown in Figure 6.1. As it can be seen, disturbances reappeared in calculating the temperature values.
Figure 6.2: Time-resolved soot temperatures at $P = 20$ atm, $r = -0.35$ mm and HAB $= 6$ mm.

### 6.3 Soot Volume Fraction

Soot volume fractions were obtained based on a time gate width with the least disturbances (20-80 ns after peak signal). The soot volume fractions as a function of radial position were as in Figure 6.3 for 10, 20, and 40 atm flames measured at 6 mm HAB. The annular soot distribution in the flame can be seen for all pressures. Results for 40 atm did not show the smooth trend of pressures of 10 and 20 atm, possibly due to flame instability issues. As a function of time, soot volume fractions were found to deviate
approximately 5% from the mean values for each location within the flame.

Figure 6.3: Methane/air soot volume fractions at 6 mm above the burner exit at pressures of 10, 20, and 40 atm. Laser beam propagation is from right to left with detectors facing into the page.
6.4 Primary Soot Particle Size

To obtain meaningful trend for particle size measurements, a time gate width of 70 to 100 ns after the peak signal was used. For each location at any given location within the flame, particulate size appeared to increase linearly as a function of time within any given measurement. Soot particle diameters as a function of radial position were as in Figure 6.4 for 10, 20, and 40 atm flames measured at 6 mm HAB. A similar annular distribution can also be observed for particle size. For each step in pressure, particle size increased at each radial position. Few negative values were obtained for 10 atm measurements which were possibly due to low signal to noise ratios.
Figure 6.4: Methane/air primary particle size at 6 mm above the burner exit at pressures of 10, 20, and 40 atm. Laser beam propagation is from right to left with detectors facing into the page.
Chapter 7

Discussion

In an attempt to match results reported in the literature, high pressure measurements were done on methane/air flames. Of great interest was comparing results with Thomson et al. [9] on which work this apparatus and the fundamental equations are largely based. Also, previous measurements by the same apparatus on the same flame were cooperatively done, and reported by Cormier [22].

Comparisons of results between the current study, previous measurements [22] and Thomson et al. [9] are presented in Figures 7.1 and 7.2. Soot volume fraction and particle size trends are similar to those of Thomson et al. [9]. Soot volume fractions show significant improvement in terms of consistency with the results reported by Thomson et al.. Largest deviations from soot volume fraction results of Thomson et al. are observed at 20 atm where peak values are roughly 57 % and centerline values are only 26 % larger. At other pressures, both peak and centerline values correspond well. Discrepancies in soot volume fraction can be partially attributed to a difference in detection volume dimensions, as slightly different parts of the flame are being detected between studies. The detection
volume in this study is defined as a 50 µm × 50 µm × 1 mm vertical line, whereas Thomson et al. use a disc with a thickness of \(~100\) µm resulting in a volume of the order \(0.001\) mm\(^3\). As for primary particle diameters, while trends are similar for pressures of 10 and 20 atm, values obtained at 40 atm show more of an annular distribution compared to those of Thomson et al.. In terms of magnitude, peak values obtained are significantly larger whereas centerline values are smaller. For the peak values, a major contributor to the inconsistencies can be differences in the time gate width used for soot particle size calculations. Fundamentally, time gate widths closer to the peak LII signal result in smaller particle diameters and vice versa [10]. When any time gate width other than 70-100 ns after the peak LII signal was used for calculations, no particular trend for particle diameter was detected. As for the centerline with lower soot loading and thus, smaller LII signals, it should be noted that as time increases from the peak LII signal, the signal to noise ratio decreases and present noises distort the decreasing trend of the LII signal and the corresponding temperature values. These distortions can alter final calculations significantly.

Improvements in the measurements for soot volume fraction (compared with results in [22]) can be attributed to better alignment of excitation and detection apertures, improved shielding, PMT and circuit board repairs, adjusting the PMT to power supply connections, reducing laser power and identifying and fixing a fuel leakage. These and other factors are discussed in greater detail in the next section.

7.1 Addressing Complications

As with previous measurements presented by Cormier [22], particle size measurements at shorter decay timescales were not meaningful and, unlike the theory’s prediction, did
Figure 7.1: Comparison of present soot volume fraction results (black symbols) with those of Thomson et al. [9] (black and white symbols) and Cormier [22] (white symbols) for a methane/air co-flow diffusion flame at 6 mm HAB.

not yield smaller particle sizes compared to cases where longer timescales were used. As observed in Figure 7.2, although trends matched literature, longer decay timescales
Figure 7.2: Comparison of present soot particle diameter results (black symbols) with those of Thomson et al. [9] (black and white symbols) and Cormier [22] (white symbols) for a methane/air co-flow diffusion flame at 6 mm HAB.

resulted in calculating fairly larger peak values and smaller centerline values for primary particle diameters. However, soot volume fractions showed significant improvements.
Cormier [22] observed significantly lower soot volume fractions than present results and those of Thomson et al. which was pronounced at 40 atm. In addition to this, winglets were observed for soot volume fraction measurements which deformed the actual annular soot distribution trend. Attenuation effects were also more visible in the previous results. Several possible contributors to these errors were identified and remedied which improved result consistencies with available literature for soot concentrations.

7.1.1 Noise

Systematic electromagnetic disturbances, generally referred to as noise, has repeatedly been reported as the greatest systematic experimental error [21, 22]. The primary cause of this interference is referred to as Q-switch noise. This disturbance is caused by high electric current peaks during lasing from both the laser’s head and power supply unit. Although generally not prevalent, it can still pose large errors to small signals when the amplitude of the interference is comparable to the magnitude of the LII signal. This contributes to a flame’s profile having greatest relative error at its lowest sooting points. Previously, copper mesh was used to shield the detection equipment which poorly blocked ambient light and contributed to random noise seen by the PMTs. Furthermore, its thin shielding poorly blocked the laser Q-switch noise. Cormier [22] recommended the construction of a shielding box made of mostly iron with a thickness of at least 1 mm. A steel shielding box was, therefore, designed and manufactured. The customized design of this box allows shielding of oscilloscope ports which was not possible in the previous setup. In addition to this, soldered connections to the PMT circuit boards which had caused the plastic wiring case to melt were rewired to be less exposed and more organized. These changes resulted in decreasing random noise. However, the oscilloscope still records small random noise signals which will be addressed in the upcoming sections. It should
be noted that complete noise elimination is not feasible since a hole on the shielding box is a necessity for the imaging of the flame onto the detection aperture.

7.1.2 PMT and Circuit Board Repairs

As mentioned in Section 5.2.3, the PMTs’ response voltages should have an increasing linear trend. Previously, during the calibration procedure, a decreasing trend for the 692 nm response voltage was observed where increasing the gain voltage resulted in lower response voltages. This abnormal trend led to sending the PMT to Artium Inc. and having it inspected and fixed. Once the PMT was sent back, calibration procedures were redone and the response voltage trends appeared normal. An interesting and dramatic change to the PMT response voltages was that, after the repair, for a given gain voltage, responses from the 692 nm PMT were larger in value than those of the 440 nm channel. This behavior is consistent with the black body radiation curve which implies that for a fixed temperature below 4000 K, the intensity is higher at a wavelength of 692 nm than a wavelength of 440 nm.

7.1.3 PMT to Power Supply Connection Adjustments

Each PMT requires a mainline ± 15 V supply. Previously, due to incorrect connections of the PMTs to the power supply, neither of the PMTs were receiving the required and correct voltage from the mainline. By adjusting the connections, this problem was fixed which led to significant improvements in the data collected from the PMTs.

7.1.4 Laser Power Reduction

Calculated temperatures from previous measurements showed peak temperatures as high as 4800 K which certainly led to an extent of soot sublimation which is not correctly
described in the LII equations used in the analysis. Generally, intense laser radiation (causing possible sublimation) can change the properties of soot, resulting in structural and morphological changes which can invalidate the soot parameters that are used in analysis equations. For LII-based particle-size measurements, it is advised that sublimation be avoided. For LII particle sizing, low fluence, with a 1064 nm excitation source, typically means \( < 0.2 \text{ J/cm}^2 \) [6]. Therefore, the laser power was turned down using the half wave plate and the resulting temperatures as shown in Figure 6.2 were inspected to stay below 4000 K. In turning down the laser power, it should be noted that, while preventing soot sublimation, the signal to noise ratio should not go below a certain value.

7.1.5 Fuel Leakage

In previous experiments, due to an unnoticed fuel line leakage, the fuel flow rate dropped leading to decreased flame height, specifically at pressures higher than 10 atm. At 40 atm, a 25 % drop of the visible flame height went unnoticed. This shortened height of the flame led to major inconsistencies with the part of the flame that was being studied. Further investigation led to identifying and fixing the leak which resulted in a 9 mm tall flame at all pressures.

7.2 Errors

As with any other experimental method, LII suffers from numerous random and systematic error sources. Many random sources can be negated through averaging and most systematic sources are accommodated through PMT calibration and scaling optical properties to match results to accepted values. Averaging reduces the effect of random error while calibration produces a scaling constant, thereby negating systematic errors.
7.2.1 Theoretical Errors

Many assumptions and simplifications used in the LII theory lead to errors which are difficult to quantify. Assuming soot particles to be blackbodies which are completely spherical and touching at single points, neglecting aggregation effects on heat absorption and conduction and considering constant values for thermal and optical properties of soot are a few of the many simplifications used for ease of theory implementation. In reality, the understanding of the physical mechanisms involved in the irradiation of soot particles and their intrinsic properties is far from being satisfactory [40]. These uncertainties lead to theoretical errors which can significantly affect measurements.

Soot Morphology

In theory, aggregates of primary soot particles are considered as an individual primary particle with an equal mass. This simplified assumption is based on the premise that aggregates are groups of spheres touching at single points and all shielding effects caused by aggregation are ignored [43, 47–50]. This is clearly in error as large aggregates do not heat evenly and do not emit radiation representative of their mass due to shielding effects. An attempt to quantify this error finds it to be significant [65].

Additionally, applying the emission spectrum of blackbodies to soot particles is predicted to result in some error.

Optical and Thermal Properties of Soot

As seen in Equation (3.4), soot volume fraction is inversely proportional to the soot absorption function. Therefore, uncertainties concerning the dependency of this parameter on wavelength and temperature greatly affect concentration measurements. Attempts
to estimate this essential property of soot as a function of wavelength have proven highly inconsistent even in the sign of the slope of the dependence [16, 41, 42, 66–69]. Studies on the function’s dependency on temperature have also had the same problem [2, 47, 70].

Furthermore, the calculated primary particle diameter based on the temperature decay rate is proportional to the accommodation coefficient [16]. As with the soot absorption function, attempts to determine an accurate thermal accommodation coefficient have proven inconsistent [16, 39, 71–74]. Although generally represented as constant, at the very least it is thought to be a function of temperature [39, 75, 76].

To accommodate uncertainties, these parameters are typically used for fitting soot volume fractions and particle sizes to physically-measured data via physical sampling and electron microscopy [6, 77]. If used as fitting parameters, much of any systematic error in the calculations will be concealed.

Local Gas Properties

At each measurement position, the local gas temperature is required, both directly and indirectly, to calculate the particle size. The local gas temperature is not only explicitly used in the particle size equation, but it also determines the heat capacity and thermal conductivity of the local gas which are used in the same equation. Errors in these values will be increased by errors in the local temperature.

Although the adiabatic flame temperature was used for all measurement positions in a flame, the temperature differs throughout the flame. Assuming constant temperature throughout the flame results in up to ± 35 % error [58]. For this reason, having a diagnostic method, that is able to measure temperature and work parallel to the LII, is
advantageous. Thomson et al. have used a numerical model based on a high pressure diffusion methane/air flame [78] to obtain the local gas temperature for LII calculations [79]. A numerical model presented by Charest [80] is available which can be integrated in the current LII analysis software for increased accuracy.

### 7.2.2 Experimental Errors

**Noise**

Ambient light, which mostly originates from the target flame’s luminosity, is one of the main contributors to random noise. This and similar noises are subtracted from measurements in post-processing, however, and do not impose significant noise to measurements. Other biased noises occurring at any point within an LII measurement can be nearly eliminated through averaging of several hundred signals during the measurement.

In contrast, as mentioned in Section 7.1.1, systematic electromagnetic disturbances are considered the greatest systematic experimental error and are primarily caused by the Q-switch noise. This disturbance contributes to a flame’s profile having the greatest relative error in cases where the amplitude of the interference is comparable to the magnitude of the detected signal. This usually happens at a flame’s lowest sooting points or after a certain time has passed from the peak LII signal. Despite using a steel box for shielding the detection equipments, the effect of the systematic noises can still be identified in the measured parameters where the soot concentrations are relatively low, specifically, at lower pressures.

**Burner Control**

Burner stage drift was observed on the order of 5 µm per minute during experiments at high pressures. The translational stage also has a resolution of ± 2.5 µm, contributing
about a 5 % error in measurement position.

**Flame Stability**

All flow controllers should be calibrated before each experiment and flow rates should be monitored to ensure constant, unperturbed flow conditions during measurements. Flames are usually smaller than usual when the pressure in the chamber is increasing. Once the pressure is fixed, the flame remains stable and the flame height should be measured using a high resolution CCD camera to ensure consistent flame size between experiments. Shorter flames generally indicate fuel leakage at a point along the system.

Since flames at high pressures are generally very small, profile measurements are quite sensitive to small instabilities. Additionally, with a spatial resolution of 50 $\mu$m, instabilities in a flame result in the wrong part of the flame being measured, which can lead to an infinite level of error. Generally, near the peak sooting region of the flame, where soot volume fraction gradients are highest, soot volume fraction can change by several factors within a small distance. At pressures over 30 atm, the methane/air flame was observed to become less stable. Outlier data points and perturbed trends are attributed to the flame fluctuations.

To overcome this error, which is considered the greatest random experimental error, an averaging of several hundred LII signals is suggested. This is in attempt of negating small fluctuations in the flame. Even with a large number of signals, this averaging may not account for all instabilities because fluctuations are likely to be far from symmetric.

**Optics**

The optics in the apparatus require precise alignment and setup to avoid error. However, perfect alignment and focus of flame images on apertures are not possible. Specifically, with a detection volume of 50 $\mu$m × 50 $\mu$m × 1 mm and the apertures mounted
on two separate tables, errors caused by micrometers of misalignment can alter measurements. There is some error in finding the correct vertical measurement position (HAB) in the flame as well. This effect is more pronounced in smaller flames.

Some minor misalignments are accommodated considering the lenses depth of field which allow for less alignment sensitivity and greater measurement accuracy. Additionally, systematic flaws in optical components of the detection apparatus are accounted for by calibrating the system. This includes wavelength-dependent differences in the transmission properties of optical components as well as imperfect focus.

Calibration

Although calibration accounts for many potential systematic flaws, it contains error itself. Linear interpolation of calibration curves for the 440 and 692 nm channel coincide with $R^2$ values of 0.87 and 0.93, respectively.

Attenuation

Although not very powerful, LII measurements are subject to attenuation effects. When shooting the laser to heat the side of the flame furthest from the excitation optics, the laser beam must pass through the entire flame before heating the detection volume. Some of the energy from the laser will be lost due to absorption and scattering from the soot in the flame. A high-sooting flame will therefore display higher attenuation effects than a low-sooting flame. This attenuation is detectible through differences between peaks of soot volume fraction at a given height.
In reality, the LII signal from every point in the flame undergoes attenuation effects which leads to some error in measuring soot volume fraction. In short, the LII signal has to pass through half of the flame to reach the detection optics and thus, some of the light is attenuated by absorption and scattering from the soot in the flame. This attenuation is not significant and is usually neglected.

7.3 Recommendations

In an attempt to improve results, the following suggestions and changes can be made to the apparatus:

Due to unterminated connections, the cable between the PMT housing and the oscilloscope is a high impedance antenna which is prone to electromagnetic noise pickup. To solve this issue and reduce error, it is recommended to attach a commercial transimpedance amplifier directly to the BNC output of the PMT module and then connect the output of that to the oscilloscope [81].

Perfect alignment of the excitation and detection aperture is complicated by the fact that they are mounted on separate tables. Currently, due to height issues with the tables, the excitation table is raised to the maximum extent and the detection table is lowered to the extent that its weight is barely on the legs and mostly on the wheels. This factor adds to the detection table’s instability. A table, designed for both the excitation and detection equipments to be mounted on, can substantially resolve vertical alignment issues which eliminates errors caused by misalignments of the detection volume. Alternatively, a rigid connection between the two LII tables would also ease alignment to some extent.
Integrating numerical models of the local gas temperature at each point in the flame into the LII analysis software can increase calculation precision. A numerical model of the local gas temperature for methane/air flames has been made available by Charest [80].

Replacing relay lenses with lenses of larger diameter would increase the amount of light detected, thus increasing dynamic range and S/N ratio.
Chapter 8

Conclusion

An LII apparatus has been fully customized for high pressure combustion diagnostics. Measurements were done on laminar diffusion methane/air flames at pressures of 10, 20 and 40 atm.

Soot volume fraction trends and values agree well with literature findings. Despite similar trends, particle size values were found to be consistently larger than values reported in literature. The differences may be attributed to the noise dominance, specifically at positions with lower soot loading, as well as larger timescales with smaller signal to noise ratios.

Overall, the measurements were mainly successful. The apparatus and analysis software can be used for soot volume fraction measurements on any laminar flame. However, particle size measurements can improve by following recommendations in regards to noise reduction and possibly software editing.
References


REFERENCES


