DEVELOPMENT OF A LASER-INDUCED INCANDESCENCE SYSTEM

by

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The ability to accurately measure solid particulate levels in various applications ranging from engines to laboratory flames has become very important in the past few decades. A new approach to measuring soot levels called laser-induced incandescence was investigated. An apparatus was designed and built in order to measure soot levels in an atmospheric laminar diffusion flame with the intent of conducting proof-of-concept measurements. The apparatus utilized highly focussed optics while collecting time-resolved data using fast PMTs which allowed measurement of both time and spatial domains. Although noise and other technical problems proved to be a concern, measurements with reasonable agreement with published results for temperature (2800 K) and the primary particle soot size (6.3 ± 2.5 nm) were achieved within the flame. Noise issues with the apparatus prevented accurate soot volume fraction measurements from being obtained. Numerous suggestions have been made as to how to improve the experiment for future use, potentially in a high pressure environment.
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1 Motivation

The study of combustion has come a long way since scientists first started pondering the nature of flame. Everything from the most basic kerosene lanterns to the most complex internal combustion engines of today’s cars utilize fire to operate and an understanding of the mechanisms involved is crucial for improving designs. In many modern combustion systems, soot plays a very significant role, for good or for ill, and there is a great deal of active research currently being conducted on this seemingly simple substance [1].

Soot has been shown to have a very broad impact in a number of different areas, some negative, others positive. Incomplete combustion which occurs in a poorly tuned car engine can lead to the release of soot particulates at the pedestrian level which will lead to an increase in respiratory ailments [2, 3]. Some studies show that soot released high in the atmosphere (i.e. from jet turbines) has a non-negligible impact on climate change [4, 5]. In addition, if soot from incomplete combustion collects on the inside walls of an internal combustion engine, the thermal absorption of the soot can cause the engine to operate at a reduced efficiency [6].

On the other hand, soot production can be a very useful thing in some applications. Heat furnaces and boilers rely on the incandescence of the soot formed inside the flame to operate efficiently [7]. Carbon black, a form of soot, is essential for products such as tires and ink cartridges [8]. Also, understanding the mechanism by which soot forms and how it evolves within the flame will lead to a greater understanding of combustion fundamentals [8]. Regardless of whether the presence of carbon is desired or despised, it can be seen that the ability to accurately measure the presence of soot in a given environment is very important.

Current commercial methods for measuring soot concentrations have been around for decades and as such they rely on older mechanical technology that has a limited scope and accuracy. Examples of such include the Smoke Number SAE test and TEM (Transmission Electron Microscope) sampling. The problem with these techniques is that they typically produce time-averaged results and it is difficult to apply these techniques to common engines without extensive modifications to an apparatus [7]. Optical measuring techniques have the potential to solve some of these problems and Laser Induced Incandescence (LII) is one of the more recent techniques developed.

The primary advantage of LII over other optical techniques (which will be discussed
later) is its versatility and sensitivity. Using LII, it is possible to obtain either time-averaged or instantaneous readings of both soot particle size and concentration within a given volume. The temporal resolution of this data is only limited by the firing rate of the laser used and can thus provide useful data in the most turbulent of flame environments. As long as the equipment is calibrated and the optics can be focused into a given region of interest (ROI), the conditions in the ROI do not matter; a burning flame or regular atmospheric conditions will both provide data. As long as soot (or any other light-absorbing nano-particle) is present, the system will be able to record an incandescent signal and with proper processing, data on the size and volume fraction can be calculated [9–11].

The sensitivity of an LII system is also superior to other optical methods of investigation. One group has managed to detect soot concentrations in a given volume of air ranging from 0.01 ppt to 10 ppm in the same experiment using the same apparatus [12]. The size of the ROI can be modified by simply changing the size of various optical components, and systems are designed to be diffraction limited so that tiny ROIs can be used for applications where high precision is necessary.

The goal of this thesis was twofold. First of all, the principles of LII had to be researched and understood; to this end a literature survey is presented later on this chapter detailing the major work accomplished by research groups around the world. Using this knowledge, a LII system was designed from scratch and then constructed inside the lab. Preliminary proof-of-concept measurements would be made using a well-characterized ethylene diffusion flame and compared to published data to ensure that the system was working properly. Ultimately, the system should be able to work with any gaseous volume including high pressure environments where soot gradients can be extremely high.

As a secondary objective, the principle of relay imaging was used to see how small of a cross-section the laser beam could realistically be focused down to. The length scales chosen for this experiment (50 µm) are at the lower reaches of what has been reported in the literature. As such, experiments with the apparatus will be able to contribute to the community at large by providing high resolution information of particle sizes and soot volume fractions within the target flame.
2 Introduction and Background

2.1 Soot Formation

2.1.1 Flame types and Burners

There are two general classifications for flame types which distinguish how the fuel and oxidizer interact. In a premixed flame, the fuel and oxidizer are mixed before an ignition source is introduced to initiate the reactions. Diffusion flames, on the other hand, have separate fuel and oxidizer regions and rely on natural convection movements caused by concentration and temperature gradients to allow combustion to occur. Regardless of the flame type, combustion only occurs within a thin cross-section of the flame known as the flame front [13]. Because the oxidizer agent in a premixed flame is evenly spread throughout the volume, the speed of combustion is controlled by the time scale of the combustion processes themselves. Diffusion flames, however, can be limited by the speed of the gas diffusion as the oxidizer molecules need time to pass through the flame front. As diffusion time scales are typically much longer than the scales of most combustion reactions, diffusion flames burn slower and with a wider flame front compared to a premixed flame under similar conditions [13]. Stabilized laminar flames provide a consistent, time-invariant flow field which can be useful for fundamental research.

Several burner types are commonly used to investigate fundamental flame properties. McKenna burners utilize a unique partial plug located above the fuel outlet to produce a thin, ‘flat’ axially symmetric flame. A metal plate can be added a few centimetres above the nozzle exit to aid in flame stability. This premixed flame is useful for diagnostics as it produces a temporally and spatially invariant flame, although a recent study has cast doubt on this claim [14]. Active cooling is often necessary to keep the base nozzle at a constant temperature to avoid a gradual pre-heating effect which occurs if the burner is left on for longer experiments.

Gülder burners are another popular burner type. These burners are configured for non-premixed flames and consist of a central nozzle for the fuel which is surrounded by a wide co-flow stream of gas. A cross-sectional design of the burner is shown in Figure 2.1. The co-flow air surrounding the fuel nozzle passes through several layers of ceramic beads before encountering the fuel. These beads remove any vorticity present in the air flow, making it easier to establish a laminar flame. The input conditions determine whether
the flame is laminar or turbulent. The laminar form is very useful as the flame that is produced is axisymmetric over its entire length.

2.1.2 Soot Formation Mechanisms

The specific mechanisms by which soot is formed is still under active study, but the most common theories are discussed below. Soot is formed within the interior of flames through various processes as the fuel is oxidized. Soot will form in a given flame, assuming that the carbon-oxygen ratio, \((C/O)_{cr}\), is achieved for a given pressure [15]. Following the initial reaction between the fuel molecules and the oxidation molecules, many different structures can form depending on how the molecules within the fuel break apart [1]. The primary precursor to soot formation has been found to be polyaromatic hydrocarbons (PAH), which are cyclic molecules consisting of carbon atoms linked together with various hydrocarbon structures on the spokes of the wheel [16]. PAH structures are formed in the initial stages of combustion where radicals and fuel remnants react to form these chain structures. For fuels such as ethane which do not have an original aromatic ring structure, the first cyclic molecule is usually formed from the reaction of vinyl and acetylene molecules via hydrogen detachment [17].
Small PAH structures require a period of growth in order to become soot particles within the flame. After the initial nucleation of the PAH structure, there are two main pathways through which the particle can grow: surface growth and particle coagulation [18]. Surface reactions occur between the primary agglomerate and PAH radicals when hydrogen detachment occurs on the surface. Both PAH radicals and acetylene are the primary growth species for this pathway as both can graft onto the open bond. These surface reactions generally increase the size of the primary particle size (approximately spherical in shape) rather than the size of the agglomerate [18]. Coagulation, on the other hand, involves larger scale collisions between larger evolved soot particles [19]. If the collisions occur when the soot is still young, the molecules stay together enough to maintain a roughly spherical shape. More mature soot collisions result in the aggregate chain structures as shown in Figure 2.2.

For laminar diffusion flames, soot inception zones initially occur within the wings of the flame where there is a high fuel concentration [13]. Soot particles increase in size by the above methods as they rise through the flame and as such, soot concentrations will have a radial dependence that also changes with the flame height. An example of the soot concentration gradients is given in Figure 2.3.

Figure 2.2: Mature soot particles in a chain for an atmospheric laminar diffusion flame. Physical grating techniques were used to collect these soot samples [20].
Figure 2.3: Soot concentration profile for an ethylene fueled laminar diffusion flame at a height of 42 mm above the burner tip. LII was used to obtain these measurements [21].

2.2 Current Measurement Techniques

Many different techniques have been developed over the years in order to determine various properties of fire. Spectroscopy methods can be used to determine the presence and concentrations of minor gaseous species but for this particular study only the techniques that can be used to measure solid soot particulates will be examined. These different measurements techniques are important to the current experiment as the LII signal must be calibrated in some fashion in order to make sense of the collected data. As Will et al. showed, it is possible to try to directly correlate the LII signal from theory, but it requires many assumptions and results in a wide range of possible combinations [22]. In the past, the LII system has been operated in tandem with another technique such as Line-Of-Sight Attenuation (LOSA) or transmission electron microscopy (TEM) sampling techniques and these separate techniques are used to calibrate the LII signal. A new technique, known as auto-calibrating LII, has recently been discovered and the current experiment will be using this method [21]. A summary of these other techniques is available in the Appendix.

One particular method that becomes very important in the current project is two-colour pyrometry. This technique involves measuring the incandescent radiation produced by a hot object at two different wavelengths. Soot within laminar diffusion flames has an approximate particle diameter of 20 nm [8, 21, 23, 24]. The Rayleigh regime defines the region in which a given particle is smaller than the wavelength of light that
it emits. For a particle of diameter $d_p$ and light of wavelength $\lambda$, the criterion is given by $\pi d_p/\lambda < 0.3$. Soot easily falls within this limit for all visible light and longer wavelengths. The power emitted by a single particle of diameter $d_p$, particle temperature $T_p$ and volume $v_p = \pi d_p^3/6$, is given by equation 2.1 [21]:

$$P_p(\lambda, T_p) = \frac{48\pi^2c^2h}{\lambda^6}[\exp\left(\frac{hc}{k\lambda T_p}\right) - 1]^{-1}v_pE(m_\lambda) \quad (2.1)$$

The $E(m_\lambda)$ term is related to the index of refraction of the soot particle and is a material property of soot which will be discussed later. Using the Wien approximation ($\exp(\frac{hc}{k\lambda T_p}) \gg 1$) on equation 2.1, it can be seen that an expression for temperature can be found by measuring the emitted power at two different wavelengths of light as follows.

$$\frac{P_p(\lambda_1)}{P_p(\lambda_2)} = \frac{\lambda_2^6E(m_\lambda 1)}{\lambda_1^6E(m_\lambda 2)}\exp\left[-\frac{hc}{kT_p}\left(\frac{1}{\lambda_1} - \frac{1}{\lambda_2}\right)\right] \quad (2.2)$$

If it becomes possible to accurately measure the intensity of light at the two different wavelengths, the temperature of the particle can be calculated. The value of $E(m)$ must be known for each wavelength band that is being used, but there is still a great deal of uncertainty in its value. For the temperatures and wavelengths associated with LII, the use of the Wien approximation introduces negligible error in the temperature calculation [25].

### 2.3 Laser Induced Incandescence Background

The general principle of LII is relatively straightforward as it involves looking at the temperature decay of the individual soot particles to determine the concentration and primary particle size. However, the actual methods involved can be very complex and there are many experimental issues that need to be considered. The study of LII has been going on for a little more than a quarter century and a great number of advances have been made over the years. New experimental methods as well as improved modeling methods continue to be needed in order to reduce the current errors still associated with this method [8]. One additional advantage of LII is that since it relies on incandescence, a property not unique to soot, it can be used to detect other nano-sized particle concentrations as well. Kock et al. uses it successfully to measure iron particles and Daun uses it for general aerosol bodies [26, 27]. See the Appendix for a brief discussion of the original theory of LII and the history of its development.
There are two different ways of obtaining the LII signal: spatial LII and Temporally-Resolved LII (TR-LII). The former uses a CCD camera to obtain snap-shot data of the entire flame, allowing position-dependent data to be collected. The majority of researchers in the past have used this method, but it does not allow time-dependent data to be obtained which is where more interesting environments can be examined. As such, a summary of spatial LII is provided in the Appendix and the TR-LII technique used in the experiment is discussed below.

2.3.1 Temporally Resolved LII (TR-LII)

Time resolved LII uses a slightly different equipment package compared to spatial LII to allow the experimenter to record the entire lifetime of the LII signal. A sample signal is shown in Figure 2.4

The advantage of recording the entire time history of the LII signal is that it allows the experimenter to resolve both the soot concentration, $f_v$, within the region of interest, as well as the effective primary particle size, $d_p$. The full theory as to how this is accomplished is discussed below, but basically the magnitude of the LII signal allows the soot volume fraction (SVP) to be calculated for the given volume. The temporal decay curve of the particle temperature allows the primary particle size to be calculated [24]. In most experiments, without smoothing algorithms, the temperature profile begins as very smooth before developing S/N issues later on as the particles cool. This is primarily
due to the fact that accurate temperature measurements require acute differentiation between short wavelength and long wavelength incandescence. Due to the rapid cooling of the particle, this is a short-lived process, and thus noise becomes predominant at later times. There is also the unavoidable background noise from the PMT which eventually takes over and drowns out the incandescent signal [8, 23, 28]. As a result, any analysis of the curve usually comes from early times where the curve is well defined. Doing so also has the advantage of not biasing against smaller particles which will cool much faster than their larger counterparts [27, 29, 30].

It is important to note that in TR-LII, the particle size, $d_p$, that is calculated from the slope is the weighted average of all the particles within the volume of interest. A great deal of theory is required in order to successfully interpret the curve [1, 29], especially since there is still debate over the size distribution of the soot particles [27, 31–34]. In general, most experimentalists and modelers have assumed a log-normal distribution typical to most aerosols [8, 25, 27]. TEM experiments have shown that the distribution varies between monodisperse and polydisperse, primarily depending on the local temperature of the gas [25, 27, 35, 36]. Some modelers have used a monodisperse assumption to temporarily make the theory simpler and then add in a correction to reduce the effect that this assumption has on the calculated size [29]. A recent study by Liu et al. show that the monodisperse assumption is valid as long as soot particles are in the free-molecular regime which occurs in most flame environments [25]. Polydisperse effects become increasingly important as the particles enter the transition regime which primarily occurs when lower temperature gases are used [25].

Another problem which arises when calculating $d_p$ is that the TR-LII theory is an ill-posed problem. There exists a wide band of values of $d_p$ and $\sigma_p$, the standard deviation on the distribution of particle sizes, any of which can be used to simulate the measured temperature decay curve [25, 27, 37]. The recent review conducted by Daun et al. shows a method of reducing the spread of possible values by simultaneously solving a size dependent function with a temperature dependent function [27]. Daun et al. also examines the effect the uncertainty in the soot parameters have on several common models today.

As the LII signal rises from baseline to maximum in about 5 ns and takes less than 1 ms to fully decay again, fast detection equipment is required. Photomultiplier tubes with a rise-time on the order of 1 ns are typically used [20, 27, 38, 39]. Michelsen et al. has utilized streak cameras, which have a response time on the order of picoseconds, to record the LII signal, but are prohibitively expensive for most applications [40]. Due to the very
short time scales involved, LII can easily be used in highly turbulent environments as shown by Boiarciuc et al. and Kock et al. [39, 41]. In highly turbulent environments however, the background noise as well as sources of noise from the experiment itself become significant due to the fact that repeatable measurements are not always possible.

2.4 Laser Profiles for LII

In order to obtain an accurate LII signal, the shape of the incident laser beam must be known. Using simple optics it becomes possible to shape the default laser beam into a desired shape, however a number of practical challenges limit how much change can be accomplished. The laser profiles used by most LII researchers can be broken into 3 categories, displayed graphically in Figure 2.5.

In addition to physical concerns, the shape of the laser profile also determines how fluence-dependent the resultant LII signal can become. These differences are due to sublimation effects which can occur as the overall power of the beam is increased. The details of these so-called ‘wing effects’ such are described in the next section, but the general fluence patterns can be seen in Figure 2.6.

2.4.1 2D Gaussian

A 2D Gaussian profile is the natural output of a TEM-00 laser and is often used for simplicity reasons [42]. The inherent problem with using an unmodified Gaussian profile is that the soot particles experience a wide range of energies which vary spatially through the beam, i.e. the particles in the centre of the volume experience a much higher energy
The LII signal response changes depending on the shape of the irradiating laser beam. The data represents theoretical modeling completed by Bladh et al. for a 532 nm excitation laser [29].

Fluence cross section of two hypothetical Gaussian laser beams and the wing effects that form. The laser power required to induce sublimation and for notable contributions to LII are both shown.

It is possible to account for this effect in the analysis but it adds another layer of complexity to the experiment [29, 44]. Also, as one increases the fluence of the laser, so called ‘wing effects’ are very noticeable when using a Gaussian profile. This effect is shown visually in Figure 2.7.

As can be seen in Figure 2.7 with a more powerful laser, a larger cross-section of the particles within the beam gain enough energy to provide a significant contribution to the LII signal. However if the laser power is increased enough to cause sublimation in the centre, seen in the top shaded portion of Figure 2.7, the optical absorption of the soot changes with the presence of the gaseous soot particles. Not only is the size of
the original particle changing with the sublimation but the gaseous particles shield the original particle resulting in a reduced LII signal [23,29,36,38].

However, with 2D Gaussian profiles, this effect has a counter-balance. As the laser energy is increased to cause the central region to experience ‘hole burning’, the power level at the edges of the laser increase and begin to contribute to the LII signal [24,38]. The net effect, as shown by Bladh et al., is that the LII signal varies linearly with fluence as seen in Figure 2.6 [29].

2.4.2 1D Gaussian

The 1D Gaussian profile is an easy profile to achieve and offers a few advantages over the normal Gaussian. This profile, seen in Figure 2.5, is achieved with the aid of a cylindrical lens which expands the beam so that the power profile is almost uniform along one axis while still maintaining the Gaussian profile along the other direction. Many researchers have used this beam as, with proper beam orientation, it becomes relatively simple to measure the beam width directly because of the constant energy along one axis [21, 45]. The Gaussian wing effects mentioned above are minimized, resulting in a longer plateau region where the LII signal is approximately constant with increasing laser fluence [29].

2.4.3 Top-hat Profile

In an ideal situation, a two-dimensional perfect top-hat profile would be used for LII experiments [7, 8]. The LII signal produced from such a shape is fluence dependent as shown in Figure 2.6. However, the beam power can be controlled so that the entire irradiated area experiences the same fluence level. As a result, as long as one operates in a flat portion of the curve (near the maximum LII signal), this is a minor influence. The fluence dependence comes from the fact that once the sublimation threshold has been reached, typically $\sim 0.3$ J/cm$^2$ for 1064 nm [8,23,38,41] excitation or $\sim 0.2$ J/cm$^2$ for 532 nm excitation [20,40,46], the endothermic sublimation processes become more important and there are no wing effects as explained above. Top-hat profiles are advantageous especially in low-fluence LII where high soot temperatures can be reached without having to worry about the extra problems that sublimation can cause when analyzing data.
Creating Top-Hat Profiles

In practice however, top-hat profiles are difficult to come by due to optical difficulties (discussed below). Several groups have tried to solve this problem by placing an aperture in the middle of an expanded Gaussian beam to sample the central, more uniform region [37, 40, 46–48]. This can be done, but without correct telescopic optics, and a laser with a low enough divergence to satisfy the Lagrangian invariant (discussed later), diffraction can still become an issue [49]. Using telescopic optics, Michelsen et al uses a pin-hole aperture to produce a 0.1 cm profile that is flat to within 20% [40]. Lee et al. uses a cylindrical lens combined with a rectangular aperture to produce a 4 cm by 100 µm profile at the measurement location [46] while Snelling et al. [45] used a cylindrical lens and pinhole to produce a tiny 36 µm x 100 µm x 100 µm ROI.

Another potential way to produce a top-hat profile is to use a multimode laser which produces a relatively uniform output, but it does not have a very stable propagation (see section 2.9.2). Investigation showed that no groups are currently experimenting with multimode lasers for LII. A third method that has been recently demonstrated is to use a custom diffraction lens which has a spatially varying index of refraction. This specific pattern on the lens allows a focused Gaussian beam to be transformed into a very stable top-hat profile of any size or diameter for a particular location [50,51]. These diffraction lenses have some very important benefits, namely the beam stability and ease-of-use. However, they are expensive (∼$8000 USD each) and they have a fixed focal length for which they can be used [50,52].

In most applications of TR-LII, the volume of interest is kept as large as possible to maximize the signal-noise ratio [7,20,41]. This can cause potential problems as the only signal that is collected is the net incandescence from the region, with every particle contributing based on the nanoparticles’ temperature. If flame conditions change considerably over the region of interest, the results that are recorded are distorted and are simply representative of the weighted average over the entire region [8]. Both signal strength and local variations must be taken into account when designing a system [37]. Several recent groups have attempted to define a small measurement volume to interpret very localized volumes of gas. Thomson et al. use a cylindrical lens to define a cross-section that is approximately 100 µm by 100 µm [45]. The laser profile Thomson’s volume is a 1D Gaussian and they achieve a physical resolution of 50 µm inside a high pressure flame. Lee et al. [46] utilize cylindrical lenses to obtain a 100 µm by
4 cm planar sheet and do not specify the laser profile achieved. Other groups utilizing TR-LII typically use much wider volumes on the order of millimetres to maximize the signal-noise ratio as stated above [22,53].

2.5 LII Calibration

In order to properly relate the measured LII signal to physical properties, the system must be calibrated. Research groups have traditionally used an additional measurement technique in addition to LII. This secondary measurement is assumed to provide accurate results, and the LII signal is calibrated against this. A review of how different research groups calibrate their systems is located in the Appendix. The current experiment uses a newer setup known as auto-compensating LII.

Most recently, a new calibration technique has been proposed by Snelling et al. [21]. Known as auto-compensating LII, the technique does not require any additional soot measurement tests to be conducted. Instead, a calibrated light source is used to provide a source of illumination to the detection equipment that is to be used to measure the LII signal. This light source can be provided by either an integrating sphere or by using the reflection of a mercury lamp off of a flat Lambertian surface. The integrating sphere consists of a halogen lamp and a hollow sphere lined with a Lambertian reflector and two output holes. One output goes towards a local spectrometer where the power spectrum can be measured in real time while the other output is simply a large hole which allows the diffuse light to spread outwards omni-directionally [54]. This second output is placed at the LII measurement location and the light is allowed to be collected by the detection optics in the experiment. Simultaneous measurement of both the measured voltage and local power spectra allow a calibration constant to be calculated. If an ordinary mercury lamp is to be utilized, as performed by Snelling et al., the Lambertian reflector is placed at the measurement location and the calibration signal is bounced off of the reflector [55].

Both of the two methods provide a diffuse, broad-band spectrum of visible and infrared light to the equipment. The integrating sphere is preferred however, since it has been shown that the tungsten and mercury lamps tend to lose their power calibration over a shorter period of time. If using a mercury lamp and no spectrometer is available, the original lamp calibration must be trusted. If both the power spectrum of the light being produced as well as the signal induced on the LII detection equipment are both measured, these can be used to provide an absolute calibration to the LII signal. In
order for this method to work, both this reference signal and the LII signal must be recorded at at least 2 different colours. This allows 2-colour pyrometry to be utilized in the analysis. Auto-compensating LII has received favourable feedback and several groups have attempted to implement it since its introduction to the literature [23, 39, 45].

2.6 LII Theory

In order to understand how LII is used as a diagnostic technique it is necessary to understand what is happening to the soot particles throughout the experimental timeline. The following section discusses the basic theory behind LII, and provides an overview as to how modelers aid experimentalists to properly interpret the LII signal.

2.6.1 Heat Transfer

Individual soot particles can be analyzed with relative ease; however, these individual particles are not seen very often in nature [1, 56]. When soot forms inside a laminar diffusion flame, it primarily forms in large aggregate fractal chains, consisting of anywhere from dozens to hundreds of primary particles loosely linked together [1, 24]. The chains can be very large, but because the individual particles are small (always less than 50 nm diameter) an aggregation theory called Rayleigh-Debye-Gans/polydisperse fractal aggregate (RDG/PFA) applies [8]. The RDG approximation states that for small particles within the Rayleigh limit ($\pi d_p/\lambda < 0.3$), aggregation effects are very small. Soot primary particles satisfy this condition for all visible wavelengths. Each chain structure can be considered a simple collection of \(N\) primary particles if two assumptions are made. First, that there is only point-contact between each primary particle, and secondly, that each particle can be treated as a sphere. The errors associated with using RDG/PFA approximations are small, amounting to 10% in extreme cases of aggregates with $\sim 400$ primary particles [8]. Most theoretical modelers use some variation of RDG/PFA in their analysis [24, 25, 29, 41].

Melton was the first to quantize the different heat transfer mechanisms which apply to individual soot particles: absorption, conduction, evaporation, radiation, internal energy change [57]. Melton’s initial formulation of the heat balance equation for individual soot particles is as follows:
\[ C_a q - \frac{2 k_a (T - T_g) \pi d_p^2}{(d_p + G \lambda_{\text{MFP}})} + \frac{\Delta H_v}{M_v} \frac{dM}{dt} + q_{\text{rad}} - \frac{1}{6} \pi d_p^3 \rho_s c_s \frac{dT}{dt} = 0 \]  

(2.3)

where \( C_a \) is the absorption cross section of the particle, \( q \) is the laser intensity, \( k_a \) is the heat conduction coefficient of air, \( T \) is the soot temperature, \( T_g \) is the local gas temperature, \( d_p \) is the particle diameter, \( G \) is a geometry dependent heat transfer factor, \( \lambda_{\text{MFP}} \) is the mean free path of the particle, \( H_v \) is the heat of vaporization of graphite, \( M_v \) is the molecular weight of the soot particle, \( q_{\text{rad}} \) represents radiative heat loss, \( \rho_s \) is the density of the particle and \( c_s \) is the specific heat of the soot.

The cooling terms in equation 2.3 all have different relative magnitudes in terms of how important they are to the cooling process. Radiation, despite the importance to LII analysis, dissipates orders of magnitude less heat than the other terms in Equation 2.3. As a result, radiation is usually neglected by modelers [24, 25, 58]. The evaporation term has the potential to dominate all of the others, but only if the laser fluence is above a given threshold [23, 29]. Sublimation has been difficult to model and understand, and it is only in the past few years that a working model has been proposed [29, 58, 59]. If the laser fluence is controlled so that the majority of the soot particles (\( \sim 95\% \) [28]) stay below the sublimation temperature, then this term can be neglected [25]. There is some evidence, however, that even low laser fluences of 0.01 J/cm\(^2\) might be able to locally allow for some form of sublimation to occur so this assumption must be made carefully [24].

At the same time that the soot particle is cooling down after the laser pulse, there are a few mechanisms which retard the cooling. The internal energy term for Equation 2.3 refers to oxidation and annealing effects, both of which are exothermic. Annealing of the soot particles occurs at much lower temperatures than sublimation and will occur at all fluence levels [24]. Oxidation of the soot will occur on the time scale of LII, however the overall effect is low [24, 60]. Although the conduction and internal energy terms dominate Equation 2.3, it is the small radiation term which is most relevant to understanding LII. An analysis of the non-radiation terms in Equation 2.3 is provided in the Appendix.

### 2.6.2 Radiation - Absorption and Emission

The interaction of soot with light forms the core of how LII is analyzed. In general, any refractive material will absorb the energy of the incoming laser pulse. It has been mentioned in discussion above that according to RDG theory, the particle is within the
Rayleigh regime for soot radiation transfer. The cross section for absorption $C_a$ from equation 2.3 is defined as follows:

$$C_a = \frac{\pi^2 d_p^4 E(m)}{\lambda}$$

where $E(m)$ is a special function based on the index of refraction of soot, designated $m = n + ik$, where $n$ and $k$ represent the real and complex components of the index of refraction $m$. This $E(m)$ function will be discussed later on. Note that in equation 2.4 there is no accounting for the shielding effects that aggregation may cause; this is because of the RDG assumption made earlier [57,60,61]. Most groups attempting to model the heating of soot simply assume that due to the high intensity of incident light, shielding effects from heating are minimal [24,25,29]. Aggregation effects are small, but noticeable, when an attempt is made to model the emission of the LII signal [24,25]. The RDG/PFA theory is an approximation, and Liu et al. and Michelsen et al. are currently attempting to develop a full simulation of the LII process [25,58,62].

Once the particle has reached the peak temperature, it begins to glow and emit radiation according to Planck’s law. The total emitted emission from a perfect black body $I(\lambda,T)$ is defined below:

$$I(\lambda,T) = \frac{2hc^2}{\lambda^5} \frac{1}{\exp \left(\frac{hc}{\lambda kT}\right) - 1}$$

where $T$ is the current temperature of the particle, $\lambda$ is the wavelength of the emitted light, $h$ is Planck’s constant, $c$ is the speed of light and $k$ is Boltzmann’s constant [7,63]. The wavelength with the peak intensity shifts to lower wavelengths at higher temperatures. This wavelength shift is used with LII to distinguish against the background flame temperature and explains why it is advantageous to heat soot up to as high a temperature as possible [24]. It should be noted that even though it is the incandescent signal that is used for analysis of the LII signal, the magnitude of the radiative cooling is orders of magnitude less than that of the other effects [30,60]. As a result, most modelers neglect this term when considering the total heat balance state of the soot particle.

### 2.7 LII Analysis

From the theory mentioned in the previous section, the LII signal can be used to calculate the soot volume fraction present within the volume as well as an approximate weighted
value for the primary particle size for the soot particles. The following analysis follows the approach used by Snelling et al. [28, 64].

2.7.1 Power Measurements and Soot Volume Fraction

The light source used to calibrate the LII system produces a power spectrum $R_s(\lambda, T_{\text{FIL}})$ which is dependent on the wavelength of light and the temperature of the filament used. If this power spectrum is directly measured with a spectrometer, the fluence of light, $P_{\text{cal}}$, that is incident on the detector optics is defined by equation 2.6

$$P_{\text{cal}} = M^2 A_{\text{AP}} \frac{A_L}{u^2} \int_\lambda R_s(\lambda, T_{\text{FIL}}) \tau_F d\lambda$$  \hspace{1cm} (2.6)

where $M$ is the magnification of the system, $A_{\text{AP}}$ is the cross sectional area of the aperture, $\frac{A_L}{u^2}$ is the solid angle of light collected by the lens and $\tau_F$ represents the total transmission spectra of the optical components (including bandpass filters). $P_{\text{cal}}$ is an absolute value with units of W/m$^3$, and the photons of light hitting the PMT induce a voltage proportional to the light intensity. The measured voltage $V_{\text{cal}}$ is exponentially dependent on the local gain value and can be theoretically represented as follows

$$V_{\text{cal}} = G_{\text{cal}} Z M^2 A_{\text{AP}} \frac{A_L}{u^2} \int_\lambda R_s(\lambda, T_{\text{FIL}}) DR(\lambda) \tau(\lambda) d\lambda$$  \hspace{1cm} (2.7)

where $DR(\lambda)$ is the detector response, $G_{\text{cal}}$ is the amplifier gain and $Z$ is the impedance of the measuring device. In both equations 2.6 and 2.7 an integral over the optical transmission $\tau(\lambda)$ and detector response $DR(\lambda)$ occurs. With the inclusion of sharp-dropoff bandpass filters, it becomes logical to simplify this integral by calculating an equivalent centre wavelength, $\lambda_c$, bandwidth, $\Delta \lambda_c$, and a peak response $\Omega(\lambda_c) = DR(\lambda_c) \tau(\lambda_c)$. The bandwidth value is defined below in equation 2.8.

$$\Delta \lambda_c = \frac{\int_\lambda \tau(\lambda) DR(\lambda) d\lambda}{\tau(\lambda_c) DR(\lambda_c)}$$  \hspace{1cm} (2.8)

The centre wavelength, $\lambda_c$, is defined as the wavelength at which the value of the normalized integration over all wavelengths of $\Omega(\lambda)$ becomes 1/2. Snelling et al. found that associated errors for using this approximation were less than 1% for LII environments using bandpass filters with a full-width at half-maximum (FWHM) value of 40 nm [21].

As stated above, particles with a primary particle size of $\sim 30$ nm lie within the Rayleigh range for conduction, that is $\pi d_p/\lambda < 0.3$. Particles with a temperature $T_p$
with the Rayleigh criterion glow and produce light according to the relation in equation 2.9

\[ \frac{P_p(\lambda, T_p)}{\lambda^6} \left[ \exp\left( \frac{hc}{k\lambda T_p} \right) - 1 \right]^{-1} d_p^3 E(m_\lambda) \]  

where \( P_p(\lambda, T_p) \) is the total emitted power in W and \( E(m_\lambda) \) is called the soot absorption function which is based on the refractive index of soot [21]. Defining the volume of the particulate to be \( v_p = \pi d_p^3/6 \) [65], the power produced per unit volume of the particulate, \( \phi_p(\lambda, T_p) \) can be defined from Equation 2.9 and is shown in Equation 2.10.

\[ \frac{\phi_p(\lambda, T_p)}{\lambda^6} \left[ \exp\left( \frac{hc}{k\lambda T_p} \right) - 1 \right]^{-1} E(m_\lambda) \]  

The particles of interest to the LII investigation are all located within the volume defined by the intersection of the excitation optics and the detection optics. The laser pulse is shaped into a cross sectional area, \( A_{\text{AP}} \) with a characteristic width \( w_b \). The final temperature, \( T_p \), reached by the particles is dependent on the local fluence level and thus the laser profile [24, 53]. From geometry it can be shown then that the experimental power, \( P_{\text{exp}} \), produced by the heated particles is as follows

\[ P_{\text{exp}} = \phi_p(\lambda, T_p) f_v M^2 A_{\text{AP}} w_b A_L \frac{u^2}{4\pi} \]  

where \( f_v \) is the soot volume present within the intersection volume and \( A_L/4\pi u^2 \) is the solid angle collected by the detection optics.

This \( P_{\text{exp}} \) will then induce a measured voltage signal, \( V_{\text{exp}} \), that can be derived in the same method as equation 2.7 as follows

\[ V_{\text{exp}} = Z G_{\text{exp}} f_v M^2 A_{\text{AP}} A_L \frac{u^2}{4\pi} w_b \int_\lambda \phi_p(\lambda, T_p) \Omega(\lambda) \tau(\lambda) d\lambda \]  

where \( G_{\text{exp}} \) represents the gain value of the PMT used in the experiment. Comparing equations 2.12 and 2.7 a large number of terms are similar. Specifically, the optical geometry is identical and will cancel out [23]. The same integral over the system transmission also appears in both and the integral itself, using the approximation in equation 2.8, can be replaced by \( \Omega(\lambda_c) \Delta_{\lambda_c} R_s(\lambda_c, T) \). From the calibration data in equation 2.7, it is possible to define a calibration factor \( \eta \) with units of V/W as follows:

\[ \eta = \frac{V_{\text{cal}}}{R_s(\lambda_c, T) G_{\text{cal}}} = Z M^2 A_{\text{AP}} A_L \frac{u^2}{\lambda^2} \Omega(\lambda) \Delta_{\lambda(c)} \]
Taking the right side of equation 2.13 and comparing it to equation 2.12, $\eta$ can be used to write an expression for the soot volume fraction $f_v$.

$$f_v = \frac{V_{\text{exp}}}{\eta w_b G_{\text{exp}}} \frac{12\pi c^2 h}{\lambda_0^2} E(m_{\chi(c)}) \left[ \exp\left( \frac{hc}{k\lambda c T_p} \right) - 1 \right]^{-1}$$

(2.14)

Every term in equation 2.14 is either a known property of soot or can be measured directly. The temperature of the soot particle $T_p$ is required, but can be found by measuring the incandescent radiation at two different wavelengths and using two-colour pyrometry. As $P_{p1} = P_{\lambda 1}\exp = V_{\text{exp}1}G_{\text{exp}1}/\eta_1$ (from equation 2.2) the soot temperature can be directly related to the calibration constant $\eta$ at the two wavelengths of interest [21].

2.7.2 Particle Size Measurement

Both spatially-resolved LII and TR-LII allow the measurement of the soot volume fraction by using equation 2.14. If time-resolved LII is used, it is also possible to calculate the primary particle sizes of the soot involved from the temperature decay curve. It should be noted that Liu et al. and others have found that this is an ill-posed problem [25,27]. Although the size of the primary particle varies throughout the flame, it is suggested that if a small enough volume is chosen, the primary particle size distribution within the given area will be monodisperse and sharply peaked around a given value [8,27].

The method used to calculate the probable primary particle size is currently based on early work completed by Snelling et al. [21]. Two-colour pyrometry can be used to calculate the soot particle temperature $T_p$ using equation 2.2. Basic thermodynamic equilibrium laws state that any hot object will cool to the surroundings and the curve will take on an exponential decay [24]. By making several assumptions (discussed in section 2.8) about the current state of the soot, it becomes possible to relate this exponential decay to the soot size.

$$d_p = -\left[ \tau_T^{-1} G\lambda_{\text{MFP}} C_p'(s) \rho_s \right]$$

(2.15)

where $\tau_T$ is the decay constant of the temperature curve, $\kappa$ is the thermal conductivity of the local gas, $\alpha$ is the thermal accommodation coefficient of soot, $G$ is the G-factor from equation A.2, $C_p'(s)$ is the specific heat of carbon and $\rho_s$ is the density of the nano soot particle.

Once the decay constant has been found from the decay curve, all other values have been tabulated and can be calculated as needed. The different material properties men-
tioned in equation 2.15 are temperature dependent with the exception of the thermal accommodation coefficient and even that has been debated [30,66]. The gas temperature within the Langmuir layer of the soot particle should be largely unaffected by the laser pulse and thus the flame temperature should be used for the calculation of $\kappa$ and $\lambda_{MFP}$ (see equation A.1). The LII signal has the highest signal/noise ratio during the laser pulse itself as well as for about 100 ns after the temperature peaks [23,67]. As such, the temperature decay is typically truncated to this high temperature region and the average measured soot temperature will suffice for the soot parameters.

### 2.8 Soot Properties

In order to properly interpret the incandescent signal from the glowing soot particles, it is essential to know several key material properties of the carbon soot. The thermal accommodation coefficient of soot, $\alpha$, is necessary for modeling the conductive cooling and affects the determination of the calculated primary particle size [8]. $E(m)$ is another important optical property that must be known. The value is a function of the complex side of the index of refraction of soot $m = n + ik$ and corresponds to the ability of a material to absorb (and re-emit) light. A third value that directly affects the precision of the LII measurement is the density of the individual soot particles, $\rho_s$. The properties of elemental bulk carbon (especially in the form of graphite) has been known for many years. However, the morphological differences between bulk carbon and nano-sized agglomerations mean that all three of these parameters differ [1,24,25,68]. In addition, the difficulty of measuring each of these properties has led to a wide discrepancy in reported values. Table 2.1 provides the range of reported values in the past few years as well as the value used for the current experiment.

**Table 2.1:** Physical Properties of Nanosized Soot Particles

<table>
<thead>
<tr>
<th>Property</th>
<th>Range of Values</th>
<th>Value For Current Experiment</th>
</tr>
</thead>
<tbody>
<tr>
<td>Soot Density $\rho_s \ (kg/m^3)$</td>
<td>1850 [69] - 1900 [30]</td>
<td>1860 [70] [21] [8]</td>
</tr>
<tr>
<td>Thermal Accommodation, $\alpha$</td>
<td>0.07 [71] - 0.9 [8]</td>
<td>0.38 [25]</td>
</tr>
<tr>
<td>Soot Absorption $E(m)$</td>
<td>0.24 [8] - 0.4 [72], $0.232 + 1254.6\lambda_d$</td>
<td>0.4</td>
</tr>
</tbody>
</table>
Liu et al. have very recently published a study re-evaluating data on well-characterized flames that have been published by other research groups [66]. As expected, the value of $\alpha$ is partially dependent upon the molecular state of the surrounding gas but they found that the proper value of $\alpha$ is 0.38 for laminar diffusion flames and other groups have agreed [24, 66, 73]. It should be noted that Michelsen has published an exhaustive numerically-derived temperature dependent value of $\alpha$ which differs considerably from Liu et al. [74]. Using carbon-nitrogen interactions as a base, Michelsen concludes with a temperature dependent data fit for $\alpha$ which results in values of 0.18-0.19 for typical LII environments with $T_g = 1900K$ and $T_s = 3500K$. The value of $\alpha = 0.38$ was chosen due to the relative agreement in the community with Liu’s work though this may change in the future.

The value of $E(m)$, which is a function of the complex index of refraction of soot, $m$, is defined by the first half of equation 2.16.

$$E(m) = IM \frac{m^2 - 1}{m^2 + 2} = \frac{6nk}{(n^2 - k^2 + 2)^2 + 4n^2k^2}$$  \hspace{2cm} (2.16)

Current research has reached a general consensus that the higher values in table 2.1 for $E(m)$ are more likely to be correct [8, 24, 58]. It should be noted that the absolute value of $E(m)$ is not necessarily required by LII; it is in fact the ratio of $E(m)$ at the two detection channels used as can be seen in equation 2.14. If the value of $E(m)$ is constant (or close enough to be approximated as constant), then the ratio simply cancels out.

There has been some support for the theory that $E(m)$ is wavelength dependent, which was a theory first put forth by Liu et al. [30] and more recently by Thomson et al. [75] and Lemaire et al. [76]. The research completed by Thomson et al. [75] shows a correlation between the relative $E(m)$ values for the range of wavelengths from 450 nm to 950 nm, which results in a 20% drop in relative $E(m)$ between the two extremes. Lemaire et al. [76] have released data showing that the ratio of $E(m)$ of 266 nm to 1064 nm is 37.5% higher than for 532 nm to 1064 nm which appears to show some sort of wavelength dependence. Both of these studies, however, are preliminary in nature. Thomson et al. [65] also conducted a numerical study which showed that having an uncertainty in $E(m)$ of $\pm 20\%$ resulted in measured concentrations varying by as much as 40%.

Another complication that arises when determining the value of $E(m)$ is that although it appears to be an intrinsic property of soot, unaffected by the mass of soot present, the morphology appears to have an affect. Experiments carried out by Thomson
et al. [75] found data that show that there is a relation between $E(m)$ and soot age. The measured $E(m)$ value for young soot (soot formed while still within the flame) as compared to mature soot measured several flame lengths downstream was different. It has been postulated that the higher hydrogen content of young soot is the cause of the difference [75].

After looking at the wide range of published studies with widely varying results, a constant value of $\alpha = 0.38$ was chosen for the current experiment. The apparent wavelength dependence has not been entirely verified by the LII community and a constant $E(m)$ value greatly simplifies the calculations.

The value of the density of the aggregate soot particles, $\rho_s$, is another property that is still undergoing some debate in the community. The density of bulk carbon varies from $2090 \text{ g/cm}^3$ to $3150 \text{ g/cm}^3$ (graphite sheets versus diamond) so it is no surprise that newly formed, flame forged soot also has different properties [77]. As Table 2.1 shows, the difference is only a few percent, but the uncertainty introduces a systematic error into the accuracy of the LII system. The chosen value of $1860 \text{ g/cm}^3$ is based from the most current models developed for soot formation [58,65] as well as recent results of TEM sampling [11].

\section*{2.9 Optical Theory}

\subsection*{2.9.1 Laser Fluence}

Any refractory particle that interacts with the laser will begin to absorb the energy and heat up. The mechanism by which the particles absorb the incoming light is dependent upon both the laser wavelength and the particle diameter, and is characterized by the Rayleigh limit defined above in section 2.6.1. RDG theory states that the particles absorb and reach a maximum temperature extremely quickly, on the order of nanoseconds [24,30]. Any interior temperature discrepancies disappear even faster, on the order of femtoseconds [24], resulting in particles with a uniform maximum temperature immediately after the initial laser pulse.

The actual temperature that these soot particles reach is dependent on the local fluence level. As long as the soot particles do not reach the sublimation point for various carbon species, the temperature will increase monotonically with increasing laser fluence [78]. The actual sublimation point for each particle is dependent upon the molecular
structure of the carbon and it typically varies between 4200 and 4800 K [8].

In general, it is the first three species (C\textsubscript{1} to C\textsubscript{3}) that are most prevalent in a sooting flame [24], so the maximum temperature of the soot particles should be kept below $\sim 4000$ K to avoid sublimation effects. Previous experiments have shown that sublimation effects are typically seen at laser fluence of $0.2$ J/cm\textsuperscript{2} for 532 nm excitation and $0.3$ J/cm\textsuperscript{2} for 1064 nm [23, 24, 59]. It is important to note that 'hot-spots' in the laser profile can lead to some particles being heated far more than other particles in the volume. This inconsistency effect can be accounted for by measuring the laser profile and calculating an equivalent width of the laser beam [21].

2.9.2 Laser Theory

In order to properly shape the laser beam into the desired fluence profile, one has to obtain an understanding both of how a laser operates and how the emitted light propagates through space. A summary of the underlying theory of laser operation is provided in the Appendix as a reference, but some relevant sections related to multi-mode laser operation are described below.

The physical design of the laser cavity defines what reflection modes are allowed to grow within the gain medium. The solutions to the wave equation of these modes are Hermite polynomials of varying orders and they define the laser’s longitudinal and transverse modes of operation. The light waves oscillating back and forth have both a transverse and a longitudinal mode. The zeroth order solution, also known as TEM-00, to the wave equation is a Gaussian profile. The TEM-00 mode has a primary wavelength related to the material as well as the cavity dimensions. Any wavelength that is not an even factor of the cavity length will not form as this violates the boundary conditions of the cavity. This condition defines the allowed longitudinal modes of the laser [79].

The transverse modes created are dependent on the geometry of the laser cavity and the spatial variation in pumping light. Most modern lasers have a gain medium that is designed to create a greater population inversion along the optical axis [80]. This leads to the Gaussian mode being preferred above all others. Other modes are inherently more lossy than the Gaussian solution [80,81]. Figure 2.8 shows the spatial appearance of the higher order modes.

Based on the orientation of the mirrors on either side of the laser cavity, one can either create a stable or unstable resonator with the state being determined by the following
Figure 2.8: Physical representations of the solutions of the transverse wave equation inside a laser cavity. Lower order modes have a higher probability of occurring [80].

\[ g_1 = 1 - \frac{L}{R_1}; g_2 = 1 - \frac{L}{R_2} \tag{2.17} \]

where \( L \) is the laser cavity length and \( R_1 \) and \( R_2 \) are the radii of curvature for the two mirrors [81]. Stable resonators occur if the product \(|g_1 g_2| \leq 1\) and represent the fact that all light with a particular vector is trapped within the gain medium. Unstable resonators allow the possibility of leakage after a few reflections have occurred. The end result of this choice is that stable resonators primarily allow one longitudinal mode of resonance within the laser cavity, typically designated as the TEM-00 mode [82]. Unstable resonators may allow more than one transverse state to exist simultaneously [82]. After a suitable build-up time, the reflection of the light saturates the gain medium so that a majority of electrons are in an excited state [82]. The laser maintains this
excited state until the reflectivity of one of the boundary mirrors change. If one mirror is not 100% reflective, i.e. \( \%90 - 99\% \), then the laser operates in continuous wave (CW) mode with a constant output. If a larger amount of power is required in shorter duration, ideal for LII applications, Q-switching is used instead [83]. With Q-switching, the gain medium is allowed to saturate with 100% reflective mirrors, which induces a very large number of coherent photons to be excited via stimulated emission. The reflectivity of one of the mirrors is then reduced, either through electronic means or via mechanical switch. The sudden loss of equilibrium causes a massive output of coherent, uniform light as the electrons depopulate the higher energy states and the trapped light floods out of the laser [81,82].

Using Q-switching techniques, it is possible to obtain a much more powerful laser pulse; however, this comes with a set of side-effects which can be detrimental to experimentalists. When the Q-switch is activated, a comparatively large amount of energy is released through the laser pulse. Part of the energy is channeled into the laser pulse used in the experiment, but the process is not very efficient (10% for most lasers) and the rest of the energy is dissipated in the form of heat and other light. This other light is in the form of a short-duration (a few ns for a typical 5-7 ns pulse width) broadband pulse which originates in the laser cavity. If proper shielding is not used, then this pulse can cause a ringing effect in local electronics. The laser cavity is enclosed within a thin metal Faraday cage which helps attenuate the pulse but additional shielding is still required for many applications when interactions occur on very short time scales [82,84].

The output of a laser utilizing a stable resonator prefers the TEM-00 mode to resonate in the cavity. A simple circular aperture placed within the beam cavity is usually sufficient to reduce higher order transverse modes to near-zero, resulting in a more pure Gaussian profile [80,85]. The temporal profile of such a laser is also typically very smooth [40, 59, 79]. Gaussian beams are very useful to experimentalists as they have well defined characteristics and behave predictably as they propagate. The output power of a TEM-00 laser is limited compared to multimode lasers, as the cavity size must be limited to avoid contamination [81,82]. The following section will go into detail as to how Gaussian beams are quantized. TEM-00 lasers have been used by many different groups to investigate LII and most take the original beam and either sample the center region using an aperture or simply expand the entire beam in one direction. However, this can still produce variations in the laser profile. [23,24,73].
2.9.3 Multimode Lasers

The Gaussian output of the TEM-00 mode presents a potential problem to LII experiments. Without careful consideration, hole-burning can occur as described in section 2.4, which complicates the analysis as sublimation must now be considered. To avoid this, a top-hat profile was desired and one way to achieve this is by modifying the laser directly to create a multimode beam output using an unstable resonator cavity.

Using an unstable resonator has its own set of advantages and disadvantages. The unstable resonator allows multiple modes of reflection to exist within the cavity. This allows the user to extract more power from the laser than single mode counterparts. The presence of the multiple modes also stretches the spatial profile from Gaussian into a more quasi-top-hat profile known as a super-Gaussian. This effect is shown in Figure 2.9.

This effect can be seen thematically by super-imposing images of the various modes in Figure 2.8. The number of transverse modes ‘k’ that exist within the cavity is dependent upon the frequency gain profile of the pumping medium [81,84]. Multimode lasers would be a boon for LII experiments for spatial reasons but they have several disadvantages. First of all, unlike the fundamental Gaussian mode, these additional modes do not natu-
rally propagate coherently. That is, the individual modes can change the spatial profile drastically over very short distances \[81,86\]. A recent study has also shown that temporal stability also varies with propagation distance for all unstable resonator cavities \[87\]. Secondly, it is difficult to focus multimode lasers down to small spot-sizes, again due to the presence of the extra modes. It has been shown that the minimum beam waist increases as \(k^{1/2}\) \[42,88,89\]. Although not applicable to LII due to broadband absorption by carbon, the minimum linewidth of multimode lasers is also wider due to the presence of additional modes \[80\]. There are no groups currently using multimode lasers for LII investigations.

Fortunately, recent laser technology has improved and a multimode laser can be modified to improve the basic performance. Two of these techniques, seeding and graded-reflectivity mirrors (GRM) are discussed here. A seeded laser assembly is primarily used for lasers trying to achieve a TEM-00 profile \[80\]. A seeded laser incorporates a smaller secondary laser which is used to pump the primary laser with a wavelength of light equal to the excited state of the gain medium \[82\]. This specific pumping drastically reduces the linewidth of the resultant laser beam and also smooths out temporal distortions in the laser pulse \[82\]. Seeding is a relatively recent technology developed in the past ten years, but it has been advocated by several groups in the LII community \[23,73,87\]. Ensuring that the temporal profile of the laser pulse is smooth is very important for modelers to know in order to develop accurate models to fit the experimental data \[24,29,90\].

A laser incorporating a GRM refers to the fact that the output mirror has a very particular diffraction pattern imbedded in its surface which changes the reflectivity of the mirror as a function of radius. This is usually done by layering various materials using a masking technique \[86,91\] or by using a uniform phase-element \[92\]. The result of this is that higher order modes are diffracted into a pseudo-Gaussian profile which allows them to propagate normally through space. A GRM component also helps reduce the divergence of the beam for both multimode and single mode operation \[80\]. The spatial propagation of this beam is still not entirely stable in that it can vary with distance, but the GRM module helps immensely and the beam is considered stable for distances less than 10\% of the Rayleigh range for the beam \[86\].
2.9.4 Light Propagation and Focusing

Understanding the interaction and propagation of light is very important for a laser-induced incandescence study. Both the excitation light beam as well as the corresponding incandescent signal must be precisely controlled in order to make an accurate measurement. The light that is produced by a typical stable resonator laser has a Gaussian spatial profile and can be generally characterized by a small number of linked parameters. These attributes include the Rayleigh range, \( z_r \), the beam width, \( w(z) \), and the far field divergence angle, \( \theta \) [42,79,81,82]:

\[
\begin{align*}
    z_r &= \frac{\pi(w_0)^2}{\lambda} \\
    w(z) &= w_0 \sqrt{1 + \left(\frac{z \lambda}{\pi(w_0)^2}\right)^2} \\
    \theta &= \frac{\lambda}{\pi w_0}
\end{align*}
\] (2.18) (2.19) (2.20)

The beam width function describes the behavior of the Gaussian beam in the near field close to the beam waist, \( w_0 \), the minimum diameter of the laser beam. An important parameter within the near field is the Rayleigh range, \( z_r \), which essentially defines the length over which the beam waist applies. At \( z = z_r \), the beam has grown by a factor of \( \sqrt{2} \) over \( w_0 \). For large distances away from the beam waist, the beam appears to increase in size linearly with \( z \), and the far-field divergence angle \( \theta \) can be defined as in equation 2.20. These parameters can also roughly be applied to a multi-mode super-Gaussian beam but it becomes much more complicated [42]. Further information can be found in Milonni [81]. A relation for the minimum beam waist is relatively simple: the presence of additional modes increases \( w_0 \) by a factor of \( \sqrt{k} \), where \( k \) is the number of modes propagating in the beam [42,88,89].

In many different applications it is useful to focus a laser beam down to a small size. Unfortunately, physical limits based on the nature of light prevent the focus from being infinitely small. Diffraction and a concept known as the Lagrangian invariant both define the smallest size down to which a given beam can be focussed down to. The Lagrangian invariant is a principle that applies to the entire optical system that is being considered. A simple form of it is summarized in equation 2.21.
\[ \theta_1 \leq d_2 \theta_2 \]  

(2.21)

where \( \theta_1 \) and \( d_1 \) refer to the incoming divergence angle and appropriate size dimension and \( \theta_2 \) and \( d_2 \) refer to the outgoing light wave. The value of \( d_1 \) can vary from system to system, but the most relevant property here will either be the minimum size of the laser beam \( w_b \) or the size of any aperture in the system. This condition must be checked for every element through which the light passes through. In essence, the product of divergence and relevant spatial dimension cannot be decreased for a given system. Thus, if a small focus point is desired, the focusing angle must be increased, i.e. via a larger lens, or else distortion of the image will occur [79,82]. For a Gaussian beam, the relevant spatial dimension is equal to the size of the beam waist and thus the initial value of the Lagrangian invariant is set by the originating laser.

**Diffraction**

Photons of light, depending on how they are treated, can be considered both waves and particles [81]. One of the characteristics of photons that is specifically wavelike is that of diffraction. Photons of a particular wavelength behave differently when they encounter physical barriers, particularly those that are of the same size order as their wavelength. When a beam of light encounters a sharp edge, the light pattern beyond the edge becomes distorted due to the changing wavefront and constructive/destructive interference [79,82]. A beam of light that encounters an object of equivalent size to the wavelength of light couples with the obstruction to an even greater extent [93]. For a circular aperture blocking a light source, the result is a radially symmetric pattern known as an Airy circle. The Airy pattern has an alternating sequence of high intensity and low intensity rings formed by the interference patterns with the distance from the first maximum to the first minimum being equal to \( d_a \). This pattern defines the Rayleigh criterion and is used as an empirical relation to define the minimum size resolution of a given optical system. For a lens of focal length \( f \), lens diameter \( a \), and wavelength \( \lambda \) the Rayleigh criterion \( d_a \) is defined by equation 2.22 [81].

\[ d_a = \frac{1.22 \lambda f}{a} \]  

(2.22)

From equation 2.22 and the discussion on diffraction it can be seen that using an aperture within an optical system can cause some profile instability, especially as smaller
apertures are used. Studies have shown that the ability of any laser beam to propagate through space, TEM-00 or otherwise, is disrupted by any aperture placed in the optical path [94]. There is a solution to this however, and it involves using a technique known as relay imaging. Relay imaging involves placing a bifocal lens, or a pair of confocal lenses, along the beam axis with the two focal points on either side of the two lenses as shown in Figure 2.10

The distance between the two lenses in Figure 2.10 should not theoretically matter since the light stays collimated and spatially constant regardless of distance traveled [42, 95]. The end result of this setup is that the image profile from one focal point is recreated at the second focus. Magnification of the profile can be achieved by varying the focus distances, so that $M = f_1/f_2$ as shown in Figure 2.10, but only the focal image will be reproduced correctly. Off-focus images may introduce distortion. This method has been used by other groups to good success [21, 23, 41, 45]. The Lagrangian invariant principle and the Rayleigh criterion still apply for this lens setup, so appropriately large lenses and/or small focal lengths must be chosen to be able to resolve small aspects of any profile that is to be relayed.
3  Experimental Apparatus

3.1  Design Goals

There were a number of parameters that had to be considered when designing the apparatus. Primarily, the apparatus should be capable of accurately measuring the incandescent signal being produced for the given target area. The apparatus would be used to investigate the properties of laboratory flames and thus had to be compatible with any and all burners currently available in the UTIAS combustion laboratory. There was also an interest in making the apparatus applicable to high pressure flames. This introduced two considerations: the physical dimensions of the sealed high pressure chamber and the size of the irradiating cross section would need to be controlled. In addition to this, atmospheric measurements and the high pressure experiments would be conducted in separate rooms so a mobile apparatus would be beneficial.

3.2  Final Design

3.2.1  Support Structure

The support structure contains all of the components of the experiment and was designed to support the necessary heavy optical tables over long periods of time without degradation in the position. The underlying structure of the cart is constructed out of 60 mm aluminum cross section made by Bosch. The thick cross section was chosen to prevent potential long-term deflection from the heavy optical plates that would be placed on top. A wooden floor was added on the lower level of the cart to allow the laser cooling unit to be placed on the portable as well as the power supply and oscilloscope if desired. In practice, however, it was found that it was more convenient to keep the power supply and oscilloscope on separate tables as far away from the laser cavity as possible due to noise concerns.

The cart itself rides on 4 separate rubber caster wheels which allow the cart to be moved with little effort. When stationary however, vibration suppression and stability are important for repeatability reasons. When in place, the cart is raised on 5 metal feet, the height of which can be adjusted individually. The entire structure can then be
leveled with respect to small imperfections on the floor using optical levels. Due to its weight and high centre of gravity, small nudges can cause it to oscillate briefly. Tests showed that the cart returns to the same neutral position as soon as movement has stopped however. The current position of the apparatus in the lab was dictated by space limitations as well as the 220V power requirement for the laser. The optical platform consists of a \(600 \text{ mm} \times 1200 \text{ mm}\) table and a \(300 \text{ mm} \times 900 \text{ mm}\) table mounted side by side. The metric tables are manufactured by Thorlabs and are designed to have standard strength but have enhanced damping characteristics. It is highly recommended [67] that vibration sources should be eliminated as much as possible. The table should isolate the apparatus from any mechanical vibrations, particularly those that could come from the laser cooling unit [67]. The tables are mounted to the support structure via bolts at the tables’ 4 corners.

### 3.2.2 Excitation Optics

The LII setup consists of two sets of optics: an excitation side that shapes the initial laser pulse used to heat the soot particles, and a detection side which detects the LII signal at the designated wavelengths. Figure 3.1 shows all optics arranged on the two optical tables.

**Laser Selection**

The characteristics of the chosen laser would affect the entire LII apparatus so many parameters had to be considered. First of all, an Nd:YAG laser operating at 1064 nm was chosen. Although working with an invisible infrared laser beam would add complications, several groups had found that significant LIF signals could be induced at 532 nm [8,96]. As the LIF signals peak during the first \(\sim 50 \text{ ns}\), selecting a 1064 nm laser removes the possibility that this could interfere with the experiment.

In order to conduct high spatial resolution measurements using time-resolved LII, a very small cross section of the flame must be irradiated with the light. This spatial size is primarily controlled by the shaping optics, but the initial laser output has an impact as well due to the Lagrangian invariant discussed earlier (Equation 2.21). As the product of the spatial dimension and divergence angle can only increase as the light passes through various optical components, it is important to make the initial value coming from the laser as small as possible. There was a large variation in the divergence values of the
different lasers considered, some of which differed by a factor of 10 [85, 97], whereas the beam diameter varied by a factor of 2 or less.

The initial laser profile also had to be determined. As discussed earlier, two popular methods are to either use an expanded, apertured TEM-00 beam or to expand the beam using a cylindrical lens and deal with a 1D Gaussian profile. A top-hat profile makes the analysis considerably easier and as such methods were investigated to allow this. Instead of having to expand the beam, one manufacturer suggested using a multi-mode laser with a Graded Reflectivity Mirror (GRM) module [85]. The laser profile should be constant for the \( \leq 1 \) m travel distance between the laser and the flame. Also, sampling the centre region would yield a constant profile without the small edge drop-off that a usual TEM-00 laser would provide. A modified Surelite-II laser with a GRM cavity modification was purchased.

The laser was measured to have a laser divergence of 0.0005 radians and an initial spot size of 3.6 mm (using \( 1/e^2 \) criterion). The laser cavity has a length of 0.18 m and a laser linewidth of 1 cm\(^{-1} \). Using these values, the Rayleigh range of the beam, over
which the spot size can be assumed to be constant, was calculated to be $z_0 = 3.83$ m from Equation 2.18.

**Laser Attenuation**

The local fluence level has a significant impact on the LII signal and as a result, it is essential to be able to control the laser power directly. Parameters on the laser itself, such as the Q-switch delay and the pump voltage can be modified to change the resultant laser power. However, independently changing these variables can change both the spatial and temporal profiles of the resultant laser beam, making reproducibility difficult [85].

The interaction of the laser with the nanoscale carbon particles is independent of the state of polarization of the light and as such, another method was chosen to control the laser power. By utilizing the combination of a $\frac{1}{2}$ waveplate and thin film polarizer (TFP), the power of the laser can be attenuated by physically turning the waveplate mounted in a rotational optical mount. The initial polarization of the laser light is random, and the waveplate acts to rotate a portion of the p-polarization to s-polarization [82,98]. The TFP then reflects all s-polarized light into a nearby beam dump and allows the remaining p-polarized light to pass through. This has no effect on the relative spatial profile. Note that the $\frac{1}{2}$ waveplate is angled approximately $5^\circ$ from perpendicular to the incoming laser beam to prevent any potential reflection of the laser which could damage the laser cavity. Also, the TFP must be angled so the incident laser beam hits it at the Brewster angle of $56^\circ$ to work properly.

**Aperture Control**

Even though the laser beam has been attenuated by a large factor in order to achieve the desired fluence level of $\sim 0.1$ J/cm², the beam is still powerful enough to damage common materials. As such, ceramic apertures are recommended due to their high damage threshold [67]. A rectangular aperture shape was chosen, primarily to simplify the analysis, though this can be swapped out for another shape with minimal changes to the analysis. The size of this aperture determines the cross section of the target that will be irradiated. The micro-sized ceramic apertures found vary in width from millimetres wide down to $5 \ \mu$m [99]. A $50 \ \mu$m wide slit was chosen for this experiment. This value was chosen to attempt to equal the smallest spatial resolution reported in the literature thus far by Thomson *et al.* [45].
When choosing dimensions for this aperture, however, the Lagrangian invariant must be taken into account otherwise distortion and diffraction effects will result in a flawed profile. The profile of the beam is not affected by the $\frac{1}{2}$ waveplate, the TFP or the infrared mirror, so the value of the invariant is equal to the initial conditions set by the laser output. From the measured laser divergence and measured beam size, the initial invariant can be calculated from Equation 2.21 and results in a value of $1.8 \times 10^{-3}$ mm·radians. With the smallest dimension of the aperture given by 50 $\mu$m, the minimum divergence angle required is 0.036 radians. It should be noted that since the Rayleigh range of the laser is so large, the actual distance between the first 4 components of the system (waveplate, TFP, mirror and aperture) does not matter.

When using a very small detection volume, there is a risk that the LII signal will be tiny and prone to signal-noise (S/N) problems [67]. As characteristics in a laminar flame do not have a very strong axial dependence at the given burner height, a long vertical dimension of 3 mm was chosen to try to maximize the signal that was obtained.

A 1:1 magnification was chosen, and the focal length of the lenses is constrained by two different parameters. The first is a desire to keep the footprint of the apparatus as small as possible for portability, while the second constraint is the physical dimensions of the high pressure rig. The distance from the viewing window to the flame inside the chamber is 228.6 mm (9") and so a 250 mm focal length was chosen. Simple geometry reveals that a 1" optical lens is sufficient to provide a low enough divergence angle so that diffraction will not be a problem.

The relay lenses are achromatic doublets specially designed for high precision optical diagnostics [100]. These doublet lenses act to correct coma, spherical aberration and chromatic aberration [100]. The lenses have been mounted inside a lens tube which automatically aligns the optical axis between these two lenses. As mentioned, the distance between the lenses does not matter and this was confirmed by experiment. The lens tube is mounted on a sliding track allowing easy repositioning of the lenses.

### 3.2.3 Detection Optics

The detection optics consist of a system designed to split the measured signal into two different light channels for use in the LII analysis. The choice of optics already limits the potential contamination of light from outside sources and in addition, all detection optics are completely enclosed within a black styrofoam light-tight box. The only opening is a
2 cm wide hole facing the flame.

**Aperture Selection**

The initial components on the detection side are virtually identical to the excitation side of the burner. The only difference is that since only thermal radiation is being transmitted as opposed to a laser pulse, a metal slit was chosen instead of a ceramic. The primary advantage of the metal slit is that the machining of the aperture is much more uniform.

The intersection of the two optical axes defines the volume of interest for the LII experiment. As 1:1 magnification is used and the rectangular apertures are identical, the volume is nominally defined as $50 \, \mu m \times 50 \, \mu m \times 3 \, mm$. The tolerances provided by the manufacturer are ±5% and 10% for the metal and ceramic plates respectively [99], resulting in a total theoretical inspection volume of $7.5 \pm 0.84 \times 10^{-12} \, m^3$.

**Light Filtering**

A single 15 cm achromatic lens collimates the light from the second aperture and this light passes through a short-wave pass (SWP) filter. The filter has a cutoff wavelength of 530 nm, and all wavelengths lower than this value pass through unperturbed. Longer wavelengths are reflected at 90°. Note that the reflection is not completely wavelength independent and this effect must be taken into account during the analysis. This SWP mirror was chosen over a typical, cheaper beam splitter as the splitter would attenuate the light signal by ~ 50% on both channels. As the signal from the small intersection volume is small as it is, choosing a splitter would only decrease the S/N.

In order to properly analyze the LII signal, the temperature of the soot is required. Two-colour pyrometry is used and thus two wavelength channels were selected for the calculation. The two channels are nominally centered on 440 nm and 692 nm with a FWHM of 40 nm for both. The centre values were chosen to primarily avoid the primary wavelengths with which $C_3$ swan bands fluoresce (473, 516, 563 and 618 nm). Shorter wavelengths are advantageous to allow easy discrimination against flame radiation, but having the wavelengths farther apart improves the accuracy of the two-colour pyrometry technique [8, 24].
Measuring the LII Signal

The current experiment investigates time-resolved signals and as such, fast detectors are required in order to fully resolve the lifetime of the LII signal. Currently, CCD cameras cannot resolve the decay curve with a high enough resolution, so photomultiplier tubes (PMTs) are used. Initially an in-house circuit was designed with off-the-shelf PMTs purchased from Hamamatsu. Unresolvable noise issues were discovered (discussed in section 5.2) so another solution was sought.

Artium Inc. manufactured two custom circuit boards with PMTs matched to the designated wavelengths. The circuit boards were taken from their own commercial LII detection system [101] and feature noise filters, op-amps and a differential output which can help with the analysis if the entire package is put together. The presence of op-amps, combined with the compact nature of the board, partially solved the antenna effect. The boxes were then completely enclosed within a 6.4 mm thick conductive box which eliminated any noise being produced by the detection electronics themselves. The power supply and gain control for both PMTs was provided by a single quad-output power supply. Mainline voltages of +15 V and -15 V were provided to both PMTs and a variable gain between 0 and 5V is manually controlled. The current setup has both PMTs receiving the same gain voltage. The circuit board electronics boost this by a factor of 200 to provide the high voltage the PMTs require.

The collimated LII signal passes through a focusing lens just before passing through the optical filters mentioned above. The focal point of the second lens is located just inside the glass surface of the PMT. Relay imaging ensures that the signal measured by the PMT is identical to the light produced in the intersection region in the flame. The location of the light beam on the measuring surface of the PMT should not affect the measured signal, but measurements showed a small deviation (∼ 10%) in signal response if the PMT surface was moved around in a 1 cm radius of gyration. However, as long as the orientation of the optics is not disturbed, the auto-calibration method used in this experiment takes this into account. A new calibration would need to be performed if the optics are disturbed.

In order to measure the fast signals that are being produced by the PMTs, a highly responsive oscilloscope is required to store and analyze the signal. For this purpose a LeCroy Wavesurfer 64Xs was used. The oscilloscope has a maximum sampling rate of $2.5 \times 10^{9}$ samples/s which is able to keep up with the 0.8 ns rise time of the PMTs. The
frequency bandwidth of 600 MHz allows all 4 input channels to be utilized if necessary. The channels of the oscilloscope were connected to the output of the Artium boards using shielded commercial BNC cables. In order to minimize potential antenna effects, the original lengths of the cables were cut in half and new BNC adapters were soldered in place. Another channel, connected via shielded cable, continuously monitored the gain voltage used to power the PMTs to observe any potential abnormalities in the signal. It is important to note that due to the short time scales involved with the LII signal, the path length that the signal travels becomes important. The signals must be recorded at the same time with reference to the original laser pulse, and any difference between the path lengths would cause the analysis to be flawed. The velocity factor of the BNC cable used is 0.75, which resulted in a temporal offset between the two signals of about 4 ns for every metre in path difference.

3.2.4 Burner Setup and Control

The current experimental setup uses a laminar diffusion flame at atmospheric pressure to calibrate the system. Once calibration has been completed on this well-characterized flame, the system can be used in any environment.

Gas Delivery System

Pure ethylene gas is used as the fuel for the current experiment. A pressurized gas tank provides the fuel to a pressure regulator which steps the pressure down to 5 atmospheres. A mass flow-rate controller provides a constant flow of gas to the burner. The flow conditions were set to be equal to those used by Snelling et al. [21], with a ethylene flow rate of 3.2 cm$^3$/s. The fuel flowing through the system exits at the nozzle of a Güldö burner with an inner diameter of 10.9 mm. The nozzle was surrounded by a co-flow system of diameter 110 mm, wherein pressurized air was allowed to flow upwards at a rate of 4700 cm$^3$/s. The air flow is cleaned and stabilized by passing the air through several layers of sintered beads below the initial surface, as can be seen in Figure 2.1. An exhaust vent is located $\sim$ 1 m above the flame tip to provide ventilation and the flame itself reaches full stability after about 30 minutes of operation. The solenoid valves used to control the ethylene fuel heat up during use and reach an equilibrium temperature during this time.
Figure 3.2: The laser power per shot measured by a power meter placed at the same location as the ceramic aperture in Figure 3.1. As can be seen, rotating the $\frac{1}{2}$ waveplate controls the transmitted power.

Beam Diagnostic Setup

Due to the orientation of the equipment, a different setup is required in order to measure the laser profile. Burn paper is a black, glossy, photographic paper with a low damage threshold and ablates very easily when exposed to a laser beam of wavelengths between 100 and 1500 nm. The degree to which the paper burns or ablates is dependent upon the local laser fluence, and as such it can be used as a cheap, easy way to determine beam location and size. Initial tests to level the laser and observation of the beam quality were performed using burn paper at various locations within the optical path. The burn paper was placed inside a plastic bag before use; the plastic bag was invisible to the laser and acts to contains the ablated material, preventing it from accumulating on any optics.

When precise measurements of the local beam profile and laser power were required, a power meter and beam profiler were both used. The maximum fluence the laser can produce is greater than the damage threshold of the power meter, and so a QEAX-25 attenuator was placed in front of the power meter to only let a measured 24.56% of the light through.

The laser power felt by the aperture was measured by placing the power meter at the same location as the aperture in Figure 3.1, and the $\frac{1}{2}$ waveplate was rotated by hand. The results of this test are shown in Figure 3.2. As expected, the power level varies sinusoidally with the rotation state of the waveplate as per $P_t = 46.5 + 42.1 \sin\left(\pi(\theta - 86.9)/44.62\right) \pm 0.21$ mJ. The fluence at the aperture was assumed to be the same level that irradiated...
the target area in the flame due to relay imaging. Due to the small cross section of the aperture, an extremely small amount of power was being transmitted even though the local fluence of that cross section was high. As such, measuring the actual transmitted power at the burner location proved impossible due to limitations on the power meter’s sensitivity.

The beam profiler CCD has a much lower damage threshold as compared to the power meter and greater attenuation is required. The apparatus was modified slightly as shown in Figure 3.3

To measure the un-apertured laser profile, the infra-red mirror is replaced by a thick 3° glass wedge with an anti-reflective coating on the front surface. The light reflected off the front surface is $\sim 3\%$ of the initial beam power and the wedged surface prevents the detector from measuring both front and rear reflections. Three neutral-density (ND) filters with a total transmittance of $5.65 \times 10^{-3}\%$ were also introduced before the CCD chip to prevent saturation. It was discovered that the changing the angle that the reflecting wedge was set at altered the image, both in shape and intensity.

This effect was found to be due to Fresnel reflection which is a polarization dependent mechanism [82]. The different modes in the laser are slightly out of phase with one another and the anti-reflective coating interacts individually with the different laser modes, resulting in the changing profile [82,102]. Thus, to get the most accurate representation
of the true profile, the reflection must be measured as close to $180^\circ$ from the incident optical axis as shown in Figure 3.3. The beam profiler is situated so that the optical path length to the profiler is identical to the distance traveled to the burner measurement location i.e. at the lens focal length.

To measure the apertured profile, the same $3^\circ$ wedge was placed after the first relay lens and this time the profiler was set such that a path length of 250 mm existed between the light traveling from the edge of the relay lens-pair, bouncing off of the $3^\circ$ wedge before being measured by the profiler. A near $180^\circ$ reflection angle was required to avoid Fresnel mode interferences.

To accurately measure the laser profile, the Newport LBP-2 USB laser profiler was positioned as in Figure 3.3. The profiler, when connected to the PC, displays the power distribution with a variable resolution depending on the sensitivity settings on the profiler. Even with the $3^\circ$ wedge, the beam power was found to exceed the damage threshold of $1 \, \mu J/cm^2$. Table 3.1 lists the attenuation factor of the included ND filters.

<table>
<thead>
<tr>
<th>ND Filter</th>
<th>Transmission (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>NG 4</td>
<td>15.1</td>
</tr>
<tr>
<td>NG 9</td>
<td>3.42</td>
</tr>
<tr>
<td>NG 10</td>
<td>1.653</td>
</tr>
</tbody>
</table>

Table 3.1: Properties of neutral density filters included with LBP-2 beam profiler

All three filters were installed to prevent damage to the profiler and minimize saturation. In order to get a stable image to appear on the PC software included with the profiler, several settings had to be changed. The ‘trigger’ value was set to 30%, and the sensitivity of the CCD was set to the ”*3/50” averaging setting. According to the documentation, this setting takes 3 readings every 50 frames and averages them together. Profiles were taken over a period of time and analyzed with the software included with the profiler to obtain information on the transmitted peak width and local distribution of power. The absolute power measurements obtained from the profiler are not as useful due to the uncertainty in the reflection coefficient of the glass wedge.

After obtaining the view of the unapertured laser beam, the same combination of wedge lens and profiler is moved beyond the relay lenses. The total path length between the profiler CCD and the second relay lens (including the $3^\circ$ wedge) is set to 250 mm so
that the image viewed by the profiler is the same image that will be irradiating the flame. The same procedure is followed as above with variable trigger settings depending on the laser settings. Note that as the profiler sits on a flat base, it is assumed to provide a good reference axis and was used to ensure that the aperture stayed vertical throughout the experiment.

The shape of the output laser profile is dependent upon the orientation of the output mirrors within the laser cavity. Distortion of the laser profile could be repaired by changing the orientation of the rear mirrors inside the laser head. If the screws are adjusted too far one way or another, the laser profile becomes distorted and stretched resulting in significant power being directed off-axis. This can result in damage to the laser itself, and potentially to the sensitive profiler. Burn paper was used as a coarse measurement before the profiler was introduced for fine corrections.

3.3 Design Methodology

The following section details the process by which the LII signal is collected.

3.3.1 Calibration Procedure

The auto-calibration technique described by Snelling et al. requires a known source to provide light to the detection optics [21]. The experimental setup is detailed in Figure 3.4.

Proper calibration is essential for the LII to work properly. For the given experiment, a SPH-6-2 SphereOptics integrating sphere (Serial # 3925), was used. Two primary measurements were recorded during this process: 1) the PMT’s optical sensitivity over the spectrum of interest and 2) the PMT’s response to varying gain voltages. The spectral transmittance of all optical components was found as well.

An integrating sphere consists of a halogen lamp providing light to a spherically-shaped Lambertian surface with two exit holes: a large 3.8 mm diameter hole for light emission and a fibre-optic coupling. The sphere was set up such that the 3.8 mm diameter exit plane was positioned at the burner location as in Figure 3.4. The lamp was brought up to a steady current of 4.166 A over a period of 2 minutes to minimize thermal shock on the system. 1 mm diameter optical fibre connected the integrating sphere to a local spectrometer which was in turn was connected to the LeCroy oscilloscope. The software
interface of the spectrometer, called SMS-500, was installed on the oscilloscope. The integrating sphere was allowed to stabilize at this amperage for 20 minutes before any measurements were taken. During this time, the spectrometer reported continuous data on the power output of the integrating sphere. Initial tests with the sphere showed that the spectrometer was reporting a very noisy and sinusoidal spectrum even though the lamp was only 10 hours into its 50 hour maintenance cycle. Correspondence with SphereOptics revealed this was a calibration issue and they provided a new calibration file which produced smoother and more realistic spectra.

All components were allowed to reach thermal equilibrium before starting and all other light sources were turned off. The gain voltage of the PMTs was set at a given value (measured by the oscilloscope) and a 300 point average was taken of the output signal from the PMT. The real-time power spectrum of the integrating sphere, $P_{\text{cal}}(\lambda)$ was captured and saved by the SMS-500 software package. Several samples were taken of this spectrum but deviations were minimal. The gain voltage, $G_{\text{cal}}$ was adjusted in 0.1 V increments from 2.5 to 3.0 V, representing the probable regime over which the LII measurements would be taken. The PMTs saturated with any higher input voltages but further experiments with ND filters were deemed unnecessary. At each interval, the PMT was allowed five minutes to stabilize (the circuit boards reached $\sim 60^\circ$C due to power

Figure 3.4: The integrating sphere provides a stable broadband source of light for calibration. The burner was removed from the stand and the sphere was mounted in its place during calibration.
Figure 3.5: PMT response to integrating sphere operated at a constant 1.446 V. The PMTs saturate at a maximum signal of 1.341 V.

collection) and the output voltage was recorded for both channels. The spectrum of light supplied by the integrating sphere was measured before and after the test to ensure a drift had not occurred during the test. An exponential function was fit to this data to provide a scaling function for various voltage inputs and is seen in Figure 3.5.

As can be seen, an exponential dependence on the gain voltage is clearly seen from Figure 3.5 and using the given relationship, any gain can be used for experimental purposes.

All optical components in the experiment have a wavelength dependence and this was found by looking at manufacturer specifications. Because the output of the integrating sphere is scaled to a single value, \( V_{\text{cal}} \), the relative transmittance of the wavelengths within the 40 nm wide bandwidth at 440 and 692 nm is important, not the absolute transmittance. All relevant data curves were multiplied together and then normalized to the highest peak resulting in a wavelength dependent value for the total transmittance spectra of the optical components, \( \tau_F \), from Equation 2.6.

### 3.3.2 LII Setup

**Alignment**

Before the start of the experiment, the alignment of the laser and optics was checked. The laser beam itself is infrared and therefore invisible, but burn paper, an infra-red viewing card or the battery-operated infra-red camera were used to trace out the laser
position at key locations. The primary method used for alignment checks was to operate the laser in single-shot mode and place burn paper on the aperture mount and at the burner location. After marking the location of the laser in both cases, a red diode laser was mounted in front of the laser at the same height as the laser beam. The diode was then adjusted so that the diode beam also hit the two burn marks which ensured that the diode was aligned with the original laser beam. The optics were then adjusted using this new visible light source. Lenses were also cleaned at this time.

**Burner and Flame**

Once the optics have been suitably aligned, the burner system was engaged. The initial co-flow pressure was set to 8 bar and the regulator back-pressure for the fuel was set to 80 bar. The air vent was also turned on at this time. The burner side of the fuel regulator was set to 3.4 bar and the electronic solenoid valves were engaged. Using the electronic mass flow controller, the desired mass flow was set by changing the percentage of maximum mass flow. A fuel flow of 3.23 cm$^3$/s was desired for the current experiment, which corresponded to a reading of 12.30% on the flow meter. The burner was lit and the equipment was allowed to stabilize for 30 minutes. The coflow air can then be increased to 28.3 L/min (or 10 CFM on the rotometer scale).

**Translation Stage**

The translation stage must be configured as well to allow the location of the flame to be found. The translation stage was connected to a computer with MATLAB installed via two serial ports, which must be linked to COM1 and COM4 ports. Both the LeCroy 64XS oscilloscope and the local lab desktop qualify but the processor on the oscilloscope is not fast enough to collect data and run MATLAB concurrently, so the lab computer was used. The control software was run using a GUI, originally programmed by Katie Bohan [56]. The two axes of movement could be controlled by either setting a desired location or by inputting a displacement value. Testing showed that the system responded better to shorter displacement requests as opposed to simply setting a farther end location. The software would crash if a distance larger than 3 cm was input. A zero position was set at the beginning of the experiment and the initial location of the burner with respect to the rest of the LII setup was measured using calipers.

It should be noted that the desired resolution of the experiment (50 µm) is nearing
the resolution limit for the stage and so a test was conducted to show that the control software for the translation stage is still capable of resolving these distances. The stage was ordered to move in 50 \( \mu m \) increments via the control software and the distance the stage actually traveled was measured using electronic calipers.

The distance traveled is consistent for shorter distances, as shown in Figure 3.6. The small deviations up and down appear to be primarily caused by the number of digits reported by the stage as opposed to any mechanical or software difficulties. However, the plot shows that if the stage is asked to move for long time steps in a single movement, it tends to move more than expected. As such, stage movements were restrained to a maximum of 10 \( \times 50 \) \( \mu m \). The power supply units for the translation stages became very hot over time and thus needed to be turned off every few hours; this may be a contributor to the pattern seen in Figure 3.6. The measurement location was set to be 42 mm for the initial experiments, as measured from the tip of the burner to the centre of the region of irradiation.

**Laser Startup**

Once all the previous equipment had been checked, the GRM-modified Surelite-II 1064nm Nd:YAG was then turned on. All best practices for operating lasers were followed as outlined in the posted regulations in the lab. To start the laser, the stop-gap key was inserted in the cooling unit to begin water circulation. The number of flash lamp pulses was noted to ensure that it was less than fifteen million, after which the Start/Stop

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**Figure 3.6:** Translation stage movement for a variety of input distances.
Flashlamp button can be pressed to start the warmup. The lamp was allowed to flash (without lasing) for approximately 30 minutes to allow the pockel cells to achieve thermal equilibrium. The Q-switch delay on the laser was set to 1.80 µs and the pump voltage to 1.25 kV; this voltage is equal to the free-running setting in the laser documentation, not the first listed value. The laser was run using two different operating modes, depending on circumstances. Setting the firing mode to F00 allowed single laser shots to be taken, which was primarily used for beam alignment. Using the F01 setting engaged continuous firing at 10 Hz and was primarily used for the LII data collection. Shot-to-shot stability and fluence profiles were found to be similar regardless of the mode chosen.

Oscilloscope

The LeCroy Wavesurfer 64Xs oscilloscope was also started while all other components were warming up. Due to the nanosecond time scales involved, it was essential that the two signals on the two different colour channels be recorded using the same time scale for all experiments. Two different trigger methods were investigated. With the first method, the trigger was set to activate when the blue channel (on channel 1) reached 200 mV on a positive spike. The maximum LII signal was controlled via the gain voltage to provide approximately 1 V so that the oscilloscope would record a signal on every pulse. Given the speed of the initial heating process, varying this trigger value from 150 mV to 300 mV had a negligible effect on the relative timing between the two channels.

The second method used involved triggering the oscilloscope on the output of the laser. The oscilloscope trigger was connected to the "Trig Out" port on the laser and this method also successfully recorded the LII signal. However, the shot-to-shot jitter for the laser was reported to be ∼ 5 ns, which, again due to the entire rise-time duration being of this order, could lead to complications. As a result the first triggering method was used for all experiments. The time scales on the oscilloscope were adjusted to resolve the first half of the decay curve and were set to 50 ns per division with a 150 ns positive offset resulting in the first 350 ns of the decay being recorded. The gain was varied based on location to produce a near-saturation maximum signal and voltage scales were set to provide maximum resolution. Values of 200 mV/division with a 700 mV negative offset were typically used.

The oscilloscope was programmed to collect the LII data automatically. Unfortunately although two colour channels are required for analysis, only one channel can be saved
using the automatic process. Given the current inherent noise in the system, a large number of samples is required to eliminate the random noise. A manual process is possible but prohibitively time consuming. The automatic process was started by selecting ‘Save Waveform’ from the file menu and then choosing a channel, location and filename for the current test. The ‘auto-fill’ button was then engaged causing the oscilloscope to save the given data channel with every subsequent trigger event.

Initially the PMT gain voltage was set at 2.8 V as this was sufficient to see the LII signal from at least one of the channels at all locations in the flame at a height of 42 mm. The gain was then adjusted to provide maximum non-saturation values. Typically 250-300 shots were taken at each location for each channel for averaging purposes.

**Cleanup**

Once the acquisition process had finished for each location, the equipment was shut down in a controlled fashion. The laser was turned off by first engaging the electronic shutter and then the mechanical shutter. The cooling water was allowed to run for 20 minutes afterwards to allow the laser cavity to cool down gradually. The fuel line was closed and the remaining fuel was allowed to burn off naturally. All optics were then covered with dust shields and all data was then transferred from the Oscilloscope to the main analysis computer using portable hard drives.
4 Data Analysis

To be useful, the LII signal data must be interpreted to yield practical results. As the recorded signal data represents the absolute intensity of light that the incandescing soot is producing, proper calibration allows this absolute signal to be related to the effective soot concentration inside the volume of interest. The temporal data can also be converted into the real-time temperature of the soot particles, and the decay constant for this graph can be related to the effective particle diameter. The current analysis is based largely on Snelling et al. [28] with a few minor changes to account for recent research and better known parameters. The software programs MathCAD 11, MATLAB v7.9 and OriginPro 8 were all used to process the data.

4.1 Setup

4.1.1 Parameters Used

In order for LII to be an effective measurement technique, the properties of the material of interest have to be known. In this case, the density of the soot particles ($\rho_s$), the thermal accommodation coefficient ($\alpha$), and the light absorption function of soot ($E(m)$) all must be known. Recent work has reduced the uncertainty in these values, but uncertainties of $\pm 20\%$ for the value of $E(m)$ are still being reported in literature [62]. After considering recent studies, a wavelength independent value of 0.38 was chosen. This may need to be revised in the future if wavelength dependency is proven to be correct. A density for soot of 1950 kg/m$^3$ was chosen for the analysis. The thermal accommodation coefficient of 0.38 was decided on, based on the extensive modeling work conducted by Michelsen et al. [58].

For the initial fluence, a value of $\theta = 162^\circ$ on the $\frac{1}{2}$ waveplate was used for these initial experiments. This corresponds to a calculated fluence of 0.15 J/cm$^2$. Subsequent experiments showed that this value may have been low as shown by Figure 4.1.

The shape of the data points in Figure 4.1 is very similar to published experiments, and clearly shows the wing effects discussed earlier. The initial $\theta$ selection is on the ascending portion of the data points, which means that local discrepancies in laser intensity could cause measurable temperature differences within the volume.
Figure 4.1: Effect on the maximum temperature reached with increasing fluence levels at a point 3.2 mm from the flame edge. The power ratio on the x-axis is in comparison to how close the laser is to maximum output. Higher energies were not investigated due to observed ablation damage on the ceramic aperture.

4.1.2 Analysis Procedure

The following steps can be followed in order to fully analyze the LII data. The Lecroy oscilloscope could theoretically run the analysis onboard, however, it lacks the computational power to collect data and run MATLAB simultaneously due to the unforeseen number of calculations required. A separate, faster computer is recommended for the post-experiment calculations. Each step will be explained below and the flowchart in Figure 4.2 is provided as a visual aid.

Loading Calibration Parameters

The calibration data must be loaded into MATLAB directly by reading the arrays for transmittance ($\tau_F$), the measured power spectrum, ($P_{cal}$), and the gain calibration curve ($G_{cal}$). All of these files were loaded into the MATLAB environment called IntegratingSphereData.mat. Note that all data points must be given to a common 0.2 nm resolution, and that compatiblewavelength.m may be required to produce suitable data sets. This calibration data is used by the software to calculate the RCS or conver-
Many different parameters in the LII experiment are wavelength dependent. To simplify the analysis, a single central wavelength, $\lambda_c$, and an equivalent bandwidth $\Omega(\lambda_c)$ is calculated for each channel. These values are defined in Equation 2.8. To obtain the total transmission $\tau$, the transmission profiles for all wavelength dependent optical components were multiplied together with the quantum efficiency of the PMT for each channel. The resulting function could then be used to calculate $\lambda_c$ and $\Omega(\lambda_c)$. This net...
Figure 4.3: Wavelength dependence of the optical detection equipment. The transmission coefficient represents the relative fraction of each wavelength of light that passes through the collection optics from the flame to the PMTs. The centre wavelength of the channels were calculated to be 439.5 nm and 689.1 nm with a FWHM of 39.5 nm and 39.8 nm respectively.

transmission can be seen graphically in Figure 4.3.

Loading and Averaging the LII Signals

After the calibration parameters have been determined, the raw LII signals must be loaded into the computer. They must be placed into the same directory on the analysis computer; typically the directory `/MATLAB/work/LIAnalysis/rawdata` is used. Each set of ~200 shots for each position should be set in a different subfolder but the 440 nm channel and 692 nm channel must be in the same subfolder. From here, all ~200 data files for each channel are averaged together using simple averaging into a single data file by running `ReadData.m`. This averaging is justified by the fact that the laminar diffusion flame is steady and any changes in signal are likely due to noise or small deviations in local soot concentrations. Error in this technique can be extracted at the same time as well to determine if the averaging is valid in each particular case. In practice, with such a large number of samples, the standard deviation of all locations was less than 5% in all cases.
Figure 4.4: Sample LII signal for both channels at a point 3.2 mm from the edge of the flame. These curves are the result of averaging the 250 samples taken at this particular location but incorporate no other post-processing.

Accounting for Noise

Once a combined LII signal has been extracted for a given location, before the interpretation of the signal can be conducted, another level of filtering is required. The LII curves for each location appear to show a systematic periodic error. The sinusoidal wave in all case studies appears to have a period of 30 MHz which matches our original observations with no flame present. As the time average value of the sinusoid is zero, a corrected LII decay can be calculated by fitting an exponential decay curve to signal after the signal has peaked. This calculation was completed manually using Origin and an example curve is shown in Figure 4.4.

The averaged data curves such as those from Figure 4.4, are used for the temperature calculations. The background level from the flame is recorded and then the data set is cropped, removing the constant data up until approximately 200 ns before the laser pulse is introduced. The cropped data is identical to the first 200 ns shown in Figure 4.5 i.e.
very close to constant. This removal is only performed to simplify the curve and reduce computational times.

Due to the different sensitivities of the PMTs to each wavelength, the two data sets could not be directly compared without first explicitly calculating the background flame contribution effects. The gain voltage for the PMTs was initially set so that both channels experienced a non-zero offset when exposed to the flame alone. This background offset was then subtracted from each channel so that the two LII curves could be compared directly. As it is the difference between the magnitude of the curves that matters for the temperature calculation, not the absolute measured value, this does not impact the first calculation. The code also finds the maximum temperature of the soot after being excited by the pulse, and stores this index value.

4.1.3 Temperature Calculation

The conversion from LII to temperature can be conducted by running the `ReadInputData.m` file, with input arguments for the input/output file names, the gain value used for the PMTs at that location, as well as the number of iterations required. The temperature calculation is an iterative process and the number of iterations can affect the accuracy.
of the outcome. Ten iterations were used for most tests and larger values produced no noticeable differences. *ReadInputData.m* calculates the apparent temperature history over the few hundred nanoseconds that the LII event occurs. A few nuances with the code are described in the following sections.

**Finding $\eta$**

The temperature calculation is a multi-step process. The first calculation that must be performed is finding the RCS constant, $\eta$, which is required to convert the output voltage of the PMTs into an optical power value. This constant is calculated as per equation 2.13 using the calibration values mentioned in section 4.1.2 by running the subroutine *RCSCConstant.m*. All values in Equation 2.13 are known at the centre wavelength, and the assumption is that these values apply to the entire narrow band that the filters allow through. This assumption is supported by Figure 4.3.

Figure 4.3 shows the effective collection efficiency for each of the channels which was found by combining the SWP mirror reflectance, the PMT quantum efficiency, the bandpass filters and focusing lens characteristics. As can be seen, there is a very high transmission throughout the region and an effective $\lambda_c$ and $\Omega(\lambda_c)$ can be calculated easily as per Equation 2.8.

Once an $\eta$ value has been calculated for a given orientation, the LII curves can be expressed in terms of power curves instead of voltages. It should be noted that this conversion currently does not take into the account the effect of absorption or attenuation that almost certainly occurs as the LII signal passes through varying chords in the flame. Also, these corrected curves have had the background signal subtracted, and that in order to determine volume fraction, the true background signal contribution must be calculated with the aid of a black body approximation.

**Finding the Soot Temperature**

At this point in the calculations, the LII signal has been transformed from a voltage PMT response into an absolute power value which represents the collected light that is impacting on the PMTs. Temperature curves were then directly calculated from the background data by taking the intensity difference between the two channels and solving Equation 2.2 for $T_s$. However, this value would be too low as the original calculation neglects the background flame radiation. To correct this number, the amount of radiation
that would be produced by a black body at the adiabatic flame temperature $T_{\text{gas}}$ is compared to the background subtracted intensity curves. This is then compared to the earlier calculated temperature and Planck’s law to obtain an improved temperature estimate. This is shown numerically in Equation 4.1.

$$\frac{P_{\text{back}}}{P_{\text{LII}} + P_{\text{back}}} = \frac{\exp \frac{hc}{k\lambda_{\text{LII}}T_{\text{soot}}}}{\exp \frac{hc}{k\lambda_{\text{LII}}T_{\text{gas}}}}$$

(4.1)

where $P_{\text{back}}$ is the actual background radiation from the flame and $P_{\text{LII}}$ is the background subtracted signal measured earlier, taken at a point 10 ns after the maximum pulse. The calculation initially assumes that the contribution from the background radiation is zero, and the calculation iterates until a stable value for both the background radiation and temperature has been found. Both the time dependent temperature curves as well as the corrected radiation curves are exported.

4.1.4 Particle Size Calculations

The effective particle size can be found from these curves by looking at the decay constant for the temperature. The problem, however, is that the time constant of the curve changes as time goes by, as varying particle sizes cool at different rates. The data points at longer time periods are relatively more stable than initial periods as they avoid the noise interference; however using a time constant at longer times will favour larger particles due to the fact they retain heat for a longer period of time. The temperature curves were transferred to Origin and an exponential fit was used to calculate the temperature decay constant using various regions of the plot.

A fitting approach for a given section of the graph was chosen as opposed to simply choosing points along the graph as performed by the NRC group due to the excessive noise in the system. An exponential fit should be able to average out the noise signals and produce the desired time constant. The time period between the peak and 160 ns was chosen for this calculation. This is longer than would be preferred (the NRC group used an 80 ns long sample [55]), but a long enough time scale was required to provide enough cycles of the noise waveform so that if the peak or 160 ns point occurred in a noise trough/peak, it would have a minimal effect on the calculated data. A secondary method which utilized the entire decay history of the signal, from peak to 800 ns, was also used to compare.
Once the decay constant was found from the plot, Equation 2.15 was used to solve for the effective particle size. It should be noted that since both this equation as well as the previous calculation for temperature depend on the value of $\alpha$, varying $\alpha$ can have a significant impact on this calculated value. Other values in Equation 2.15 are known to a much greater certainty. All temperature dependent variables were evaluated at 3000K.

### 4.1.5 Particle Concentration Calculations

Once the temperature history of the particle has been found, it is now possible to find the average soot concentration within the volume of flame. Equation 2.14 defines how the SVF is found and it contains only two experimentally determined variables. Both the instantaneous temperature of the soot $T_s$ and the absolute power intensity of the light calculated in section 4.1.3 are required. It theoretically should not matter which of the two channels of light (440 or 692) is used though both were used to compare against one another. It should be noted that ReadInputData.m initiates this process, but the file SootVF.m is used to actually perform the calculation and any changes to soot properties should be made in this file.

### 4.2 Results

Preliminary results were captured from a ethylene laminar flame operating from a G"ulder burner. Preliminary tests showed the LII signal to be very noisy and difficult to interpret reliably, and errors on all values are significant. Figure 4.4 shows the typical LII signal with a curve fit for the decay portion.

#### 4.2.1 Noise Measurements

The LII signal was plagued with noise very soon after the laser pulse. This noise pulse appears to have affected both the power supply and the PMTs themselves, though shielding reduced the noise produced by the PMTs. Figure 4.6 shows the voltage output of the power supply immediately after the laser pulse. The seven samples shown are a random sampling of the laser being operated for two minutes at 10 Hz.

These tests were conducted with no flame present. It should be noted that since the PMT response is exponentially dependent upon an instantaneously stable source voltage, these fluctuations have a major impact on the signal. Another issue that should
Figure 4.6: Effect of the Q-switch laser pulse on the gain voltage seen by the PMTs. The high frequency noise resonates at approximately 30 MHz, starting immediately after the laser pulse which is also shown. Ringing typically lasts for 600 ns after the pulse.

be noted is the rather significant RMS noise during the time when the voltage should be constant before the pulse. It is unknown if this noise is due to power supply switching (suggested by the laser manufacturer), random radio noise or thermal noise from the environment. This noise has an oscillation frequency of 600 ± 100MHz (limited by the measuring resolution).

4.2.2 Averaging Results

As mentioned above, the system is very prone to noise and so a basic averaging technique was applied to approximately 200 shots at a given location. Any random noise should cancel out from the averaging and thus any systematic noise will be revealed. Figure 4.5 displays the results of this procedure for a given location.

While it is clear that there is a systematic noise error in line with what was measured in Section 4.2.1 above, there also appears to be another effect in play. Both the maximum temperature reached as well as the slope of the LII pulse appear to be slightly different for many of the tests, in some locations more than others. Figure 4.7 shows the maximum voltage recorded on the PMTs as well as the standard deviation of this value after taking
Figure 4.7: Relative variation in the maximum signal recorded for all measurement locations. Maximum uncertainty occurs near the edge of the flame. The edge of the flame is shown at 5.4 mm. The deviation varies widely across the flame, ranging from insignificant to 25% of the measured signal. Note that this file simply shows the maximum voltage achieved. The power supply gain voltage was adjusted for each test so that the LII signal would stay far away from saturation (the PMT achieved saturation at 1.33 V).

4.2.3 Temperature Results

Figure 4.8 shows the calculated temperature decay for each of the 26 measurement locations chosen.

As can be seen, there is similar behavior for all positions, with the rapid rise and slow decay characteristic of the LII process. All data locations were set to trigger at the same time, so the fact that some of the curves, particularly those taken later in the testing, i.e. near the edge of the flame, appear to heat up slower resulting in the maximum temperature being achieved slightly later than earlier tests. This shift is never more than 4 ns as compared to earlier tests.
Figure 4.8: Temperature decay profiles for all measurement locations. The largest signals occur at the first few locations taken, and the signals begin to temporally stretch towards the end of the data set. Each line represents a single location within the flame.

4.2.4 Particle Size Results

The effective particle size was calculated as described above for each of the different locations. As mentioned earlier, choosing which part of the decay curve to take the particle size from has a large difference in the calculated particle size. To demonstrate, two such methods are displayed in Figure 4.9.

As can be seen, there is a factor of two difference between the two methods. The error bars shown in Figure 4.9 come from two primary sources: $\alpha$ and $\tau$. The uncertainty in $\alpha$ contributes a basic 25% uncertainty to the size value, and this tends to dominate the calculation. In some of the locations in the flame, however, the signal was especially noisy, even with averaging completed, and thus the uncertainty in $\tau$ is comparable to the uncertainty in size.

4.2.5 Particle Concentration Results

Once the instantaneous temperature of the soot particles was known, the soot volume fraction could then be calculated as per Equation 2.14. All values aside from the temperature, $T_p$ and the signal strength $V_{exp}/\eta$ are known and as such the comparison between
Figure 4.9: Particle sizes as calculated from the temperature decay. Results from two methods are shown: Using the temperature curve from peak to 160 ns after, and from peak to 720 ns.

these two values should reveal how much soot is present. Unfortunately, the noise signals shown in Section 4.2.1 proved particularly problematic for this calculation. Specifics will be discussed in the next section, but in essence the fact that this calculation calls for the LII signal from a single channel to be used increases the effect of the electronic noise. As a result of this noise interference, no reasonable measurements of the soot volume fraction could be found at this time.
5 Discussion

As the data in the previous chapter indicate, the apparatus currently provides data which are reasonably in agreement for soot temperature and soot size but the noise signals have prevented an accurate measurement for the soot volume fraction [21,39,45]. The typical radial wing profile, as seen in Figure 2.3, is also difficult to see in these experimental results. This section will be divided between discussion regarding the results themselves as well as current (and past) issues with the experimental apparatus which will need to be resolved.

5.1 Data Discussion

5.1.1 Initial Setup

All the physical apparatus pieces appear to be working properly, but Figure 4.1 should be noted. The shape of the curve is similar to the fluence plots in most published data [8], though it currently appears to follow the curve from a Gaussian fluence distribution, i.e. continuously increasing LII with fluence. The initial fluence level at which the experiment was performed is on the initial rise. This could introduce errors in temperature due to unequal distribution of fluence in the volume. Future tests should be performed at $P/P_0 \sim 0.35$ to minimize this effect. Future experiments could also expand this plot to see any high-fluence LII dependence. It should be noted that measuring the fluence profile using the profiler at higher powers may require extra filters to attenuate the beam down to levels that the profiler can handle. In addition, at $P/P_0 > 0.5$, the laser began creating bright flashes when hitting the ceramic aperture. This is merely fluorescence but it may also be signaling ablation of the plate, which could affect the shape of the aperture at higher power levels. As long as the power output is controlled by the TFP, the spatial profile of the laser beam should not change.

5.1.2 Temperature Calculations

The temperature calculated using two-colour pyrometry is lower than expected. For the fluence level used in the experiment ($0.15 \text{ J/cm}^2$), most reported literature recorded temperatures from 3000 K to 3500 K, while the measured peak temperatures for all
locations hovered between 2800 and 3000K, as can be seen from Figure 4.8. Three potential causes for this discrepancy come to mind. One possibility is that the calculated fluence could be lower than its actual value. As discussed below, there were difficulties with using the laser beam profiler, and the calculated fluence could be based upon an inaccurate integration region size. If so, then this could be solved by increasing the power, which would raise the maximum temperature seen in Figure 4.1.

A second possibility is that since temperature is found by comparing the two channels of LII with one another, a temporal offset between the two channels could affect the calculation. If one channel has been time shifted such that the oscillation peak of one occurs more than 5 ns (the apparent peak width) after the other, the comparison could cause a lower temperature to be calculated. Careful examination of the data shows that there is some temporal variation between different locations in the flame (see Figure 4.8) but both the 440 and 692 channels are always synchronized with respect to one another in each individual location.

A third possibility is that our value of $E(m)$ is incorrect. Our initial selection of $E(m) = 0.38$ is in the centre of the accepted range of values (see Table 2.1), but there is still considerable uncertainty in its actual value, especially compared to other groups. This is likely a minor concern seeing as the temperature is logarithmically dependent upon the value of $E(m)$.

It should be noted as well that when looking at the raw LII signals in Figure 4.4, the larger signal is much noisier than the other. A single gain voltage was used for both channels, and as such a much larger signal was obtained from one channel over the other. If the gain was increased such that the 440 nm channel were closer to saturation, it began to display the same noise characteristics as the larger 692 nm channel. It would seem that operating at lower gain values would help eliminate the effect of the noise, but then one loses resolution and the ability to distinguish the proper decay constant of the curve. Separate power supply voltages for both channels may also help to solve this.

5.1.3 Soot Volume Fraction

As mentioned in the previous section, the apparatus currently has difficulty producing SVF measurements. The current SVF calculation is entirely based on the calculation procedure provided by Snelling et al. [55,67,103]. However, when the measured temperature and LII signal curves were analyzed using the MATLAB code described in Section
4.1.2, the resulting SVF values were nonphysical. The magnitude of the calculated SVF was off significantly and it showed abnormal temporal behavior as well.

After encountering this problem, every part of the analysis code was double-checked to ensure its accuracy with the work completed by Snelling et al. [21]. Although a sample data set from NRC was not available to run through the new computer code written for this experiment, the two programs appeared to produce the same results. Direct, full-scale testing of the Snelling et al. code was not possible due to the way that they had designed their MathCAD software, but small test-runs showed that both the original MathCAD software and the new MATLAB program produced the same results.

Since Snelling et al. [21] had produced accurate SVF data using that software package, it was then decided that the problem must lie elsewhere. Given the relative success of the experiment in calculating both the soot temperature and particle size, there were only a few factors which could cause such a large distortion. One thing that was immediately noticed was that for both the particle size and temperature calculations, it is the ratio of signal strength between the 440 nm and 692 nm channels that is important. It is only for the SVF calculation that the absolute signal occurs. The electronic noise has a large effect on the gain voltage (as seen in Figure 4.6) which is common to both PMTs. As a result, it is reasonable to believe that the noise impacts both channels and thus a calculation which uses the ratio of the two channels will have the impact of the noise reduced. The noise effect will not be perfectly negated, since the PMT response is exponentially dependent on the gain voltage and the amount of light hitting the PMT. However, with the SVF calculations, there is no such ratio and the full effect of the noise can be seen. It is therefore absolutely required that the noise issue be resolved before the apparatus can produce SVF data.

5.1.4 Particle Size

The effective particle size which was calculated from the data set shown before appears reasonable, though it is highly erratic. Snelling et al. who used similar conditions, reported an expected particle size of $3.8 \pm 0.1$ nm using both their LII setup and physical sampling [21]. Using a short time period (160 ns after the pulse) actually overpredicted the particle size by a factor of about 3. However, when a longer time scale was used, the data actually agrees within the calculated error for the experiment for the majority of the data points in the flame. Spatially, the particle size appears to be constant throughout
the flame, with outlier points popping up towards the edge of the flame. However, there should be distinct changes in the soot size in the oxidation regions near the edges of the flame [46]. Small changes can be seen superficially in Figure 4.9 but it is not clearly defined.

It should be noted that the difference between the two analysis methods is strange and contrary to published trends. As can be seen, the time constant in the temperature decay graphs changes as time goes on.

As can be seen from Figure 4.8, there is no single decay constant which can describe the entire cooling process of the heated soot particles. As smaller particles cool faster than larger particles, using a longer integration period should bias the calculated particle size towards larger particles. However, the current analysis shows the opposite effect. Using a shorter time period with the analysis actually gives a larger particle size as compared to the longer time period. Spot checks at intermediate intervals showed this trend to be consistent with the current analysis. It is currently unknown why this is happening but it is likely an artifact of the excessive noise. The noise signal is strongest in early times and then dies off as shown in Figure 4.6.

5.1.5 Absorption Effects

One major assumption that was made during these calculations is that the effect of absorption from soot and from the gas itself was negligible. This assumption may not be fully substantiated, and calculations should be introduced to take this into account. It is unlikely, even in a heavily sooting flame, that significant attenuation will occur within the volume of interest, due to its small size. However, the SVF of a G"ulder burner should be radially symmetric, and it is possible that, due to the orientation of the equipment, measurements may not initially reflect this. One can imagine getting different readings due to varying path lengths of the light within the flame.

5.2 Apparatus Complications

5.2.1 Noise

Initially, off-the-shelf PMTs were purchased from Hamamatsu Inc., however severe noise issues prevented them from being used for reliable experimentation. Looping power leads
acted as a large antenna which amplified any electronic signal in the environment. Binding the power leads together and surrounding the result with copper wire mesh helped slightly, but the Q-switch laser was still introducing a noise pulse that was completely swamping any signal that the PMT could detect. A significant radio noise level was also picked up by the power leads. After other attempts to solve the problem were not successful, it was suggested by the NRC research group that we look at an Artium Inc. provided solution. Artium provided the circuit boards and mounted PMTs but not their custom analysis software.

The advantage that the Artium boards provided was that all sensitive electronics and filters were located extremely close to the PMTs so that electronic noise would be minimized by the small electronic paths. Unfortunately, much of the power processing was proprietary and the specific chip and component design of that aspect of the board was sealed. Once the Artium boards were configured, they were able to almost completely eliminate the radio noise that had been picked up by the Hamamatsu PMTs in a darkened room. They also allowed the LII signal to be resolved as a smooth curve with no noticeable deviations even with the Q-switch pulse. However, the boards introduced another problem that took several weeks to notice. The noise reduction mechanism that the boards utilized included a pair of inductors which were used to remove high frequency noise from the output signal.

The LII heating process normally takes about 5-10 ns but the inductor limited the time response of the system and expanded the reported heating time to $\sim 1 \mu s$. As a result, the response curved was stretched in the time domain so that the decay curve was lasting almost $1 \mu s$. This meant that it became very difficult to interpret the actual soot temperature, as significant temperature gradients only exist for the first two hundred nanoseconds. Artium itself uses these PMT boards in their portable products which measure soot emissions over a longer time scale on the order of seconds. As such, the short term changes in temperature can be accounted for. This result is not acceptable to the current experiment since it is necessary to be able to interpret the true response curve in the proper time domain. After talking with the engineers at Artium, these inductors were physically removed and while solving this one problem, others emerged.

The inductors were extremely useful in reducing noise, and as such the modified PMTs began picking up a great deal of environmental noise. Re-wiring the power with twisted-pair cable and adding an additional copper-mesh sheath helped to reduce this, but once again the Q-switch noise was causing noise pulses that were showing up on
the oscilloscope even when no flame was present. Enclosing the PMT and circuit board completely in an electrically grounded solid metal box was the only way found to reduce this noise signal. Copper mesh was not sufficient to protect the circuitry from the high frequency component of the Q-switch noise. The end result was that a clear LII signal could be produced from the oscilloscope but one that had an additional electrical noise signal that could not be positively identified. Components were isolated to try to identify the cause of the problem with minimal success. The trigger for the noise appears to be the laser trigger as the oscilloscope clearly records the noise signal even when the PMT detector is covered. Both detectors appear to show the same noise signal (the waves are in phase and similar in magnitude), even though the cable length is slightly different (due to physical construction reasons).

One possibility is that something in the electronics was providing the source of the signal. The power supply (and connectors) are a likely candidate but the PMTs themselves may also play a part. In order for a signal to be correctly transmitted along a waveguide such as a BNC cable, both ends of the wire must appear to have the same impedance with respect to the signal. If there is a mismatch in impedance values, ringing can occur as part of the signal is reflected at the boundary instead of being transmitted to the rest of the circuit. This explanation would explain the ringing signal, though we cannot find exactly where this is happening. The oscilloscope inputs are naturally set at 50 Ω, and Artium reports that the output of the PMT boards are also set at this value. However, it is possible that removing the inductors to fix the response problem as mentioned above may have shifted the load impedance of the PMT. This seems to be a likely source for the problem although further research is required to determine this.

Another potential problem area is the power supply and its connection with the PMT. The power supply is providing a set voltage and thus is not impedance matched to the input of the PMT. A single twisted pair cable is used to provide the power, as these cables are known to be highly resistant to noise of all frequencies. The PMT and power supply are both grounded together using the building ground obtained through the power supply’s 120V power cable. Tests showed that even when the power supply was isolated from the PMT, the output voltage (as measured via the oscilloscope probe) was affected by the Q-switch of the laser. A ringing signal was found. Shielding the power supply via copper mesh helped reduce the signal but it was still present. This effect is very important to resolve since the LII response from the PMTs is exponentially dependent upon the gain voltage. Even small deviations of mV (which is the magnitude of deviations that
were measured) could have a significant impact on the output voltage. In light of this realization, the power supply was returned to the supplier for testing to ensure that there was no internal flaws with the unit. Their basic testing showed that nothing was wrong mechanically and a visit was paid to their labs to try to recreate the noise issues. Even when placing the both the power supply and oscilloscope into a faraday box with several centimeters of solid metal shielding, the power supply still recorded a small random noise signal with a very short duration of 5-10 nanoseconds.

This noise was likely coming from random radio noise in the atmosphere, but because of its low occurrence rate and duration with respect to the length of the experiment, it is unlikely that this will impact a noticeable number of the tests. This independent noise signal is a limitation on the power supply itself; 4 independently controllable power sources are required to power the PMT. It was difficult to find scientific power supplies with 4 outputs, and even the chosen power supply showed vulnerability to noise when examined at high frequencies. Discussion with engineers at the supplier, after the purchase, revealed that this is a very common problem with most power supplies, as the conversion from AC to DC produces consistent RMS voltages, but instantaneous voltages on small time scales are not as smooth. A custom power supply may be required, or high quality single-output power supplies may need to be purchased.

Another possible source of the noise was the translation stage power supply. The power supplies, it was realized later, were largely unshielded and contained large transformers. During the tests, these stages were located directly below the burner and thus fairly near the apparatus. The power supplies were left on for the entire data collection process, and were still drawing power even when the stages were not moving. As a result, they may have contributed to the general noise signal seen by the electronics. Of greater importance is the fact that the power supplies were not on when the integrating sphere was being used for calibration.

5.2.2 Laser Profiling

One of the tasks that was surprisingly challenging to accomplish during the experiment was accurately measuring the laser profile. Initially, a large quantity of burn paper was acquired to provide a qualitative representation of the beam profile. This allowed the laser beam controls at the back of the unit to be optimized in a coarse fashion to ensure that no obvious hot spots existed which could damage the beam profiler and/or power
meter. However, as the laser saturates the burn paper’s damage limit, the laser profiler itself was needed to fine tune the final alignment. A few problems were encountered with the laser profiler, however.

First of all, when first connected, the profiler was set up in transient mode to be able to interpret laser pulses and all three attached ND filters were used to prevent the profiler from exceeding its damage threshold. However, even when properly aligned, the profiler did not consistently show the laser image. Changing the power trigger settings to 40% of the maximum range (with a 1/32 sensitivity) resulted in consistent images; however, they were not stable. The image would continuously scroll upwards on the display and then repeat itself coming up from the bottom. Initially it was thought that this was a physical problem with the laser and that for some reason it was producing a scanning laser beam that was oscillating up and down since the frequency for each cycle was about 10 Hz. Subsequent tests showed that this was not the case and the software was re-configured. Averaging was turned on for the profiler (setting the timing interval to 3/50 appeared to work best) and continuous wave mode was used. This resulted in a usable image, but technicians at the manufacturers of both the laser profiler and the laser were puzzled as to why the laser pulse mode was not working.

The imperfections in the uniformity of the laser profile itself are almost certainly caused by the imperfections in the ceramic aperture itself. Micro-machining is difficult and Lenox Lasers provided the best quality-price comparison that could be found, with quality defined by the smallest tolerance in the size of the machined aperture [99]. However, diffraction patterns are particularly strong when the wavelength of light being used is close to the size of the object the light is interacting with. Looking at both apertures (ceramic and metal) through the microscope, the ceramic side wall was noticeably rough, although no imperfections were noticed in the metal aperture at 50 times magnification. The largest divots of the ceramic were measured using to be approximately 4 µm deep and they irregularly occurred along both sides of the ceramic for the entire 3 mm height of the aperture. This is likely the main cause of the unequal power distributions that are seen in the measured relay image. As the laser has a wavelength of 1.06 µm, interactions with this surface will cause airy diffraction patterns, the interaction of which will cause concentrations of high and low fluence levels.

The current calculations assume an average fluence level across the aperture, however some improvements can be made. The first recommendation is to find an alternative aperture material that has a high damage threshold, is non-reflective and can be more
easily machined to a smooth edge. The second is to replace the current 50 μm aperture with an aperture with a wider area so that although wall imperfections will still cause interference patterns, the wider aperture will mean they will cause less of an effect. In addition, a wider aperture may result in a larger LII signal and will improve the overall S/N ratio of the experiment.

It should be noted that during the course of the experiment, a second high power ceramic aperture had to be ordered to replace the original. All apertures were inspected using a microscope prior to use, and initially there were no noticeable defects in the ceramic material greater than the listed tolerance (< 5%). The original aperture however was used in the apparatus before the laser beam was properly aligned, and as such hot-spots were present and a subsequent microscope viewing showed that chunks of the aperture wall had been damaged by the laser, resulting in a very nonuniform profile being transmitted. The replacement was ordered and the laser power was lowered using the attenuator optics to ensure no further damage; subsequent observations of the aperture confirmed this.

The original power meter that existed in the lab was designed to measure wavelengths in the 500 nm range; as such, the material surface was susceptible to damage from the current laser even at reduced power levels. After some superficial damage was inflicted upon the surface when the power meter surface by itself was exposed (tests showed no difference between affected areas and unaffected regions), a specially designed attenuator had to be purchased.

A third issue that arose while testing was with the laser profiler itself. While looking at the profiler images, spherical imperfections can be seen occurring at the same locations on the CCD. These imperfections are likely caused by small dust particles somewhere, although it could potentially be a distorted CCD surface causing these. The blot persisted despite careful repetitive cleaning of all optical surfaces with propanol and optical paper. The CCD chip itself on the profiler was deemed too sensitive for solvents although compressed air was used to try to clean the surface. The profiler was positioned during all tests to try to prevent these blobs from intersecting with the ROI, but in some cases this was impossible. This caused a minor difficulty in interpreting the images, but should have a negligible effect.
5.2.3 Laser Stability and Imaging

One of the more challenging aspects of the experiment was adjusting the laser to provide as uniform a profile as possible. Linked with this was also the need to have the beam stay stable and unchanging for the entire beam path. There were essentially 4 settings on the laser that could be easily controlled by the user: vertical mirror control, horizontal mirror control, the Q-switch timing level and the voltage of the pumping cavity. Changing the orientation of the horizontal and vertical mirrors controls the shape of the lasing cavity and subsequently the image that it produces. Ideally it should be possible to orient the mirrors so that a perfect reflection occurs and they should not needed to be changed for the entire experiment. Unfortunately, it was found that changing both the Q-switch and the pump voltage had an effect on the profile and thus these would have to be re-adjusted after every significant change.

The pump voltage controls how much power is being provided to the laser cavity and thus a higher pump voltage means more laser power. Fortunately for this experiment, as long as the minimum fluence level was achieved, a large power is not required; simply control over the profile is desired. Initially, due to confusion over the documentation provided by Surelite, the pump voltage was set to 1.65 kV. Testing was conducted over a ±0.2 kV range but 1.65 kV appeared to provide the best image on a short distance scale despite some noticeable focusing effects. Ultimately, setting the voltage to 1.17 kV resulted in a much more consistent image. The reason for the inconsistent behavior noticed earlier was likely due to a thermal lensing effect. The large amounts of power being dumped into the resonator crystal resulted in a higher radial temperature profile than the manufacturer’s design specs called for. As there is a non zero $dn/dT$ ($n$ being the index of refraction) value for Nd:YAG crystal, the cavity acted as a lens which focused the light slightly; this was only noticeable over a distance of more than one metre. Setting the pump voltage too far below the design voltage has the opposite effect and caused the laser to begin to diverge from the cavity. As the power level can be controlled using the TFP and half-waveplate, changing the pump voltage can be used to control the divergence of the laser if it becomes a problem.

The Q-switch trigger controls the time that the laser cavity is allowed to build up before the Q-switch is activated, releasing the laser pulse. A constant Q-switch value of 1.80 µs was used for the entire experiment, with minor deviations to see the effect on the laser profile. In essence, from experimentation and from talking with the Continuum
representative, the Q-switch also controls the power output of the laser. A shorter Q-switch delay allows less build-up time for the laser, resulting in a lower excited population in the upper energy levels of the Nd-YAG crystal and less energy is released. A longer build-up time results in more energy being released, up to the point where saturation occurs, after which longer delays will not have an effect. Rough experimentation did not pin-point a time at which this occurs for the current laser. It should be noted that adjusting the Q-switch results in a profile change as well. Shorter Q-switch times created profiles which were slightly more uniform overall, with fewer local hot spots, as compared to longer Q-switch times, even with refinement of the image using the rear mirrors.

5.2.4 Flame Stability

Before starting the experiment, the burner and fuel assembly were tested to ensure a constant air flow and fuel flow rate so that a laminar flame would form. However, once confirming the fuel flow rate, this test was not conducted again. After the experiment had concluded, several months later, Emre Karatas began to use the same fuel delivery system and he noticed that a leak had developed in the fuel regulator which was causing a pressure drop and subsequent flow irregularities. It is possible that this may have developed between the time when the initial flow tests were conducted and the LII investigation itself. This may explain why the LII signals are so chaotic in a laminar flame. It should be noted however that the pressure drop observed by Mr. Karatas was not seen during the experimental proceedings at any point.

5.3 Primary Recommendations

Several suggestions for future experiments and changes to the experiment can be made. First of all, a larger aperture would result in easier to interpret results. Choosing a tiny aperture like this has led to several complications, which, although interesting, have made accurate proof-of-concept measurements challenging. Starting with a larger aperture should also limit diffraction effects and lead to a more controllable laser profile; once the system has proven to work the aperture size can easily be reduced for more detailed work. Secondly, a proper Faraday cage should be built around the power supply and particularly the connections to the oscilloscope where they are currently unshielded. As an aside to this, the apparatus was initially designed to be fairly compact with everything
close together. Limited testing, however, showed that spreading the equipment out may in fact reduce the noise level in the system. Thirdly, Mr. Dan Clavel has suggested that one way to improve the S/N ratio at the source is to attach a small resistor (47 kΩ) in series with the PMT output at the oscilloscope using a T-connector. This will effectively allow a larger signal to be recorded by the PMT without worry of DC saturation.

The theory of auto-calibrating LII works well and has been proven in many different labs around the world. However, in this particular situation where the LII setup is being developed for the first time and with limited experience with regards to the technical challenges, many obstacles were encountered. A second, simpler method of measuring soot concentrations, such as the LOSA setup currently in the lab or TEM sampling should be paired with the LII system to properly corroborate the results found.
6 Conclusion

The most significant challenge encountered was noise which impacted every aspect of the experiment. Even with fitted curves that tried to minimize the impact, it was difficult to consistently estimate the true value of the LII signal. The source of the noise is hypothesized to be the laser, though other sources, such as random radio noise and switching noise coming from the power supply may also contribute. Recent tests have shown that separating the equipment as much as possible also aids in reducing the noise considerably.

With regards to the data taken, however, the temperature plots agree fairly well with published results, although they are typically $200 - 300\degree K$ lower than most cited works. This may be a result of underpredicting the fluence level due to an inconsistent laser profile. The measured temperature decay also predicts the effective soot particle size within error ($6.3 \pm 2.5$ nm), as long as the entire decay curve is utilized to make this calculation. The current analysis scheme does not reliably predict the soot volume fraction. Further investigation into the calibration procedure and noise sources is required to solve this discrepancy.

Overall, the LII development can be deemed a partial success. The design and construction of the apparatus were successfully completed. The LII signal itself has been observed and measured in a laminar flame environment. Although there appears to be issues with regards to the interpretation of this signal, this is a solvable problem and, if the recommendations laid out in the previous section are followed, there is no reason why the LII apparatus will be fully functional in the near future.
A Appendix

A.1 Other Techniques to Measure Soot

A.1.1 Mechanical Sampling Techniques

Methods that fall under these categories work by physically obtaining a sample of the soot. This can be done both inside the flame as well as in the exhaust of the flame. Gravimetric sampling is one of the simplest measurement techniques. A small filter is typically directly above the flame in or near the smoke plume [104, 105]. Residence times vary but are typically on the order of several minutes [3, 105]. The mass of particles collected with this filter is then weighed and the soot volume fraction can be found directly [35]. However, this results in only mature soot being measured and thus is not as useful for time-dependent measurements.

Thermophoretic sampling utilizes a very fine metallic grid or thin surface which, is usually mounted on a pneumatic stage [106, 107]. The size of the mesh varies depending on the desired spatial resolution and number of samples but is typically on the order of several tens of µm. Thermophoretic force, a force that particles experience due to an imbalance in temperature, cause particulates to be attracted to the grating [108]. The particles that are attracted can then be viewed using transmission electron microscopy (TEM) to find out the primary particle size as well as the form that the aggregates take. Thermophoretic sampling is primarily used to gather data inside the flame itself, and the pneumatic stage controls the residence time. Typical sampling times are on the order of 100 ms [106, 107].

There are several advantages to mechanical sampling and for these reasons it is still fairly popular today [3]. The gravimetric sampling technique does not require specialized equipment aside from the filter paper; however, air contamination prevents this from being very accurate in a commercial application. An alternative measurement, known as the SAE smoke number tests, measures the optical opacity of a filter after a given residence time in an exhaust and uses this to determine emissions. However, there is an appreciably large error on this measurement which make its precision limited [3]. It also performs poorly in heavily sooting flames where the filter can become heavily clogged, which affects the flow of the plume and it cannot be used to sample the flame itself due to quenching effects [23].
Thermophoretic sampling can be useful as it provides direct physical samples of soot from specific locations within a flame, but several problems remain. Even with short residence times it is still an open question as to whether quenching affects the flame structure [8,109]. Also, flame structures change on a much faster scale compared to what a pneumatic probe can achieve, so a time-averaged sample is still being measured [13]. Lastly, it can be difficult to ever get in situ engine measurements with thermophoretic sampling because of the extensive modifications required. As a result, thermophoretic techniques are usually reserved for lab environments only.

### A.1.2 Optical Techniques

Optical techniques have many intrinsic advantages over physical sampling. In general they are very fast techniques, able to take snapshots of even the most turbulent flames [7]. They are widely thought to be non-intrusive in that measuring the soot does not noticeably affect the flame structure, though there is some debate over this theory [8,43]. Some optical techniques, such as Raman and Rayleigh measurements, take advantage of the fact that light will scatter differently based on the local density of the air [7,110,111]. Witze et al. used Rayleigh scattering to complement their LII measurements in their particular study [112]. Line-of-sight-attenuation (LOSA) takes advantage of the light absorbed and scattered by a given concentration of soot particles. LOSA uses a CCD camera to take several images of a collimated light beam with and without the presence of a flame. The images can then be subtracted and analyzed (using Abel inversion for axisymmetric flames) to determine the radial soot volume fraction within a flame [56, 78]. A similar method, laser-extinction, uses the attenuation from a laser beam to determine local soot fractions. The LOSA approach is used to calibrate the works of Kock et al., while laser extinction is used by Lee et al., Faeth et al., Hofmann et al. and Smooke et al. [46, 48, 109, 113]. Pastor et al. show that a spatially large 2D attenuation measurement provides greater calibration accuracy as compared to multiple single-location measurements [114].
A.2 Thermal Theory of Soot

A.2.1 Conduction

Conductive heat transfer takes into account the cooling process due to physical transfer of energy from the soot particles to the surrounding gases [24]. There is minimal heat conduction between particles in the fractal aggregates which form due to the fact that a) they are touching at a single point only and b) all particles within the LII volume are assumed to reach the same maximum temperature, so gradients within the particle are non-existent and small within the aggregate structure. When considering conductive heat transfer, the most important dimensionless number is known as the Knudsen number, 
\[
Kn = \frac{\lambda_{MFP}}{L},
\]
where \(L\) is the given length scale of the system, usually \(d_p\) for LII. The mean free path of the local flame environment is defined by the following equation
\[
\lambda_{MFP} = \frac{k_p T_0}{\sqrt{2} \sigma_a P_0} \tag{A.1}
\]
where \(k_p\) is the Boltzmann constant, \(T_0\) and \(P_0\) are the gas temperature and pressure, and \(\sigma_a\) is the mean molecular cross-section of the gas [24]. The true local concentration of gases around each soot particle will vary, and so for this calculation air at 1900 K is used as an approximation [24]. Using typical values for atmospheric air gives a \(\lambda_{MFP}\) of 440 nm. If \(Kn\) is very large, the particles exist in the Knudsen, or free-molecular, regime where particle-air collisions are rare and particle dynamics are defined by the kinetic theory of gases [24,25,115].

If \(Kn\) is found to be closer to unity (Michelsen uses a condition of \(Kn < 5\sqrt{\frac{\pi}{2}},\) Liu et al. use \(Kn < 10\)) then the particle resides in the transition regime where kinetic theory alone is not enough to describe the dynamics [24,25]. Dasch [78] proposed a method for dealing with the transition regime where two volumes are created around the particle. The inner layer is of radius \(\lambda_{MFP}\) and is treated as a free-molecular regime while the outer Langmuir layer is treated as completely continuum. Melton originally assumed that the large aggregate structure placed the soot particle clearly within the transition regime [57]. Recent studies have generally agreed that this transition regime only occurs in high pressure environments and that a kinetic approach is sufficient for atmospheric flames [24,29]. Some groups still include a correction that modifies the conduction heat transfer term, but the correction remains small for atmospheric applications [25,60,64,116]. This correction term appears in Equation 2.3 as the \(G \lambda_{MFP}\) term. This \(G\) factor is a geometric
A term that is defined in Equation A.2. [60]

\[ G = \frac{8f}{\alpha(\gamma + 1)} \]  \hspace{1cm} (A.2)

where \( f = (9\gamma - 5)/4 \) is the Eucken factor, \( \gamma \) is the ratio of specific heats for the local gas and \( \alpha \) is the thermal accommodation coefficient for soot [24,25,57].

## A.3 High Pressure Considerations

One of the many advantages of an LII approach to measuring soot is how tunable the measurement volume is. A smaller irradiation volume is of particular advantage when investigating high pressure flames, where all the soot reactions occur in a much smaller area. The diameter of a given diffusion flame decreases as \( P^{0.5} \), drastically increasing the soot concentration gradients and potentially affecting the soot formation mechanisms [56]. As such, a finely focused beam could be used to induce a LII signal within a small volume providing localized information about soot morphology, however several groups have identified potential problems with this method [37,117,118].

In order to obtain a clear LII signal, it is necessary to quantify the state of the beam profile as it passes through the flame. A primary assumption for LII is that the laser energy absorbed by all soot particles within a given volume is spatially constant [23,59]. With atmospheric flames, soot concentrations are low enough to be neglected in most cases [8,119], however beam attenuation becomes significant in high pressure environments as shown by recent experiments [37,117,118,120]. Ochoterena et al. reported a laser intensity reduction of 70% for typical diesel flame environments as the beam passes through the sooting region [117]. This attenuation forces the experimenter to use a more powerful beam which in turn will introduce significant soot sublimation which is complicated to model [24].

Another potential problem with extending this technique to high pressures is beam steering. The hot gases being produced by a given flame produce a gas density distribution within the flame. The index of refraction of a given gas is related to the local density and as such the beam location could change within the flame [82]. Given the small scales required to resolve the flame properly with LII, these slight changes in density cause beam steering to occur within the flame itself, resulting in a false signal as reported by Charwath et al. and Geigle et al. [118,120].
References


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