Tomographic imaging of combustion zones using tunable diode laser absorption spectroscopy (TDLAS)

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TOMOGRAPHIC IMAGING OF COMBUSTION ZONES USING TUNABLE DIODE LASER ABSORPTION SPECTROSCOPY (TDLAS)

A Dissertation

Submitted to the Graduate Faculty of the Louisiana State University and Agricultural and Mechanical College in partial fulfillment of the requirements for the degree of Doctorate of Philosophy

in

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by

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Abstract

This work concentrates on enabling the usage of a specific variant of tunable diode laser absorption spectroscopy (abbr. TDLAS) for tomographically reconstructing spatially varying temperature and concentrations of gases with as few reconstruction artifacts as possible. The specific variant of TDLAS used here is known as wavelength modulation with second harmonic detection (abbr. WMS-2f) which uses the wavelength dependent absorbance information of two different spectroscopic transitions to determine temperature and concentration values. Traditionally, WMS-2f has generally been applied to domains where temperature although unknown, was spatially largely invariant while concentration was constant and known to a reasonable approximation (±10%). In case of unknown temperatures and concentrations with large variations in space such techniques do not hold good since TDLAS is a “line-of-sight” (LOS) technique. To alleviate this problem, computer tomographic methods were developed and used to convert LOS projection data measured using WMS-2f TDLAS into spatially resolved local measurements. These locally reconstructed measurements have been used to determine temperature and concentration of points inside the flame following a new temperature and concentration determination strategy for WMS-2f that was also developed for this work.

Specifically, the vibrational transitions (in the 1.39 \( \mu \)m to 1.44 \( \mu \)m range) of water vapor (H\textsubscript{2}O) in an axi-symmetric laminar flame issuing from a standard flat flame burner (McKenna burner) was probed using telecom grade diode lasers. The temperature and concentration of water vapor inside this flame was reconstructed using axi-symmetric Abel de-convolution method. The two different sources of errors in Abel’s deconvolution - regularization errors and perturbation errors, were analyzed and strategies for their mitigation were discussed. Numerical studies also revealed the existence of a third kind of error - tomographic TDLAS artifact. For 2D tomography, studies showing the required number of views, number of rays per view, orientation of the view and the best possible algorithm were conducted. Finally, data from 1D tomography was extrapolated to 2D and reconstructions were benchmarked with the results of 1D tomography.
Chapter 1
Introduction

Efficient monitoring and control of combustion processes require quick and preferably non-intrusive methods of measurements for temperature and/or concentration of different combustion species [1, 2]. Specific examples include automotive and jet engines, industrial burners, incinerators, furnaces and also process control in process industries. Additionally, over the past few decades, due to the increasing concerns over environmental pollution, trace gas monitoring has gained importance. Polluting gases generally have very low concentrations due to the diluting effects of atmosphere and hence measurement techniques need to be extremely sensitive with high detection limits. Laser based measurement approaches are generally quick and non-intrusive and therefore have significant potential for application in such areas.

Of all laser based applications for gas detection, tunable diode laser absorption spectroscopy (also abbreviated as TDLAS) has gained significant popularity in recent years compared to others because of two primary reasons – low cost and availability of lasers. Low power (rated ~ 2 to 10 mW), semiconductor diode lasers are relatively cheaper and due to the recent advances in optical technologies in telecommunication, many lasers are available in the near-infra-red (NIR) wavelength region. Incidentally, most species of interest in combustion have active rotational-vibrational transitions in the near-IR region which can thus be probed by such lasers. All absorption spectroscopy methods are, however, “line-of-sight” (LOS) methods – i.e. signals are absorption values integrated along the beam path. TDLAS methods are hence generally applied to areas with uniform temperature and concentration of the absorbing species [3, 4, 5, 6]. Although some researchers have used TDLAS in conjunction with tomography for non-uniform domains [7, 8, 9, 10], most of these work use large number of spectroscopic transitions with direct-absorption (or DA) spectroscopy. Very few studies exist with two-transition (or 2-line) thermometry and even fewer with wavelength modulation spectroscopy using second harmonic detection (or WMS-2f), for domains with large variation in temperature and concentrations. This work, therefore, concentrates on tomographic reconstruction of temperature and concentration zones of a premixed laminar flat flame using two-line WMS-2f with “non-optimized” modulation depths.
This dissertation is divided into the following sections: a) chapter 1 provides an overview of the related work that has been done in this field and the objectives and motivation behind this work; b) chapter 2 covers the necessary theory for TDLAS; c) chapter 3 describes the tomographic algorithms in details for 1D and 2D tomography along with work done on beam arrangement optimization as well as maximization of precision and accuracy of tomography; d) chapter 4 describes the different kinds of artifacts that can be encountered during the process of tomographic-TDLAS using numerical results; e) chapter 5 describes a novel experimental setup, automation of the same and other system integration tools designed for 1D tomography as well as data reduction for finding spatially varying 2f-height distribution; f) chapter 6 contains the experimental results of 1D tomography and its extension to 2D tomography; g) and finally, chapter 7 contains the conclusions and outlook for this work.

1.1 TDLAS: A Brief Summary
Tunable diode laser absorption spectroscopy (TDLAS) is a type of laser diagnostic technique that is based on absorption of visible or infrared light by the gas through which it is traveling (Fig. 1.1). Absorption occurs at discrete wavelengths that are specific to each molecular species. The wavelength-dependent attenuation of light is a function of gas properties, which forms the measurement principle of absorption spectroscopy. The principal properties of interest for TDLAS measurements are gas temperature and species concentrations. Detectable species include both major and minor combustion species viz. carbon-dioxide (CO₂), water vapor (H₂O), carbon monoxide (CO), oxygen (O₂), methane (CH₄), acety-
Figure 1.2: Absorption bands of H₂O, CO and CO₂ in the 6000 to 7000 cm⁻¹ (1.38 to 1.66 µm) range (y-axis not to scale)

Lene (C₂H₂), formaldehyde (CH₂O), ethylene (C₂H₄) etc., as well as radicals like hydroxyl molecules (OH). Each molecular species has a spectral “absorption signature” that acts as a “fingerprint” - in the sense that different molecular species have a large number of spectroscopic transitions that absorb light at different wavelengths. These are often referred to as the absorption bands (Fig. 1.2). The two variants of TDLAS – viz. direct absorption (DA) spectroscopy and wavelength modulation spectroscopy with second harmonic detection (WMS-2f) – are both well suited for gas sensing and monitoring of temperature and species concentration in combustion [11, 12]. With the increased availability of telecommunication band semiconductor diode lasers, these methods have gained popularity as they are non-intrusive, quick, can have concentration sensitivity of the order of parts-per-billion (ppb) [1], can be adapted to probe specific gas species and since most of the rotational-vibrational transitions of the molecules mentioned above fall in the IR and near-IR wavelength range which is covered by these diodes. Both these methods (DA and WMS) probe a pair of absorption transitions which are functions of temperature and concentration but have significantly different absorption profiles due to difference in values of their characteristic
spectroscopic parameters. In DA, temperature and concentration can be inferred from the ratio of the integrated spectral absorption. These measurements are, however, highly susceptible to measurement noise due to a low signal-to-noise ratio (SNR). Diode lasers typically exhibit low-frequency, “1/f” type, intensity noise [13]; white noise contributions – typically much smaller in magnitude – are due to photodetector shot and thermal noise. By manipulating the emission wavelength of a diode laser via its injection current, wavelength modulation is used to create a modulated absorption signal and its higher harmonics, which are functions of temperature and concentration of the absorbing species. In WMS-2f, the second harmonic, in particular, is detected and used to determine the temperature and concentration of the flame. The WMS-2f technique (also known as derivative spectroscopy [14, 15]) improves upon DA by using only the high-frequency components of the modulated signal and rejecting the DC component. Since the "1/f" type flicker noise mainly corrupts the DC component of the signal which is not used at all, a significantly higher SNR is achieved in WMS.

1.2 Previous Work with TDLAS

Although the science of absorption spectroscopy has been in existence for quite some time, some of the early works pertaining to TDLAS in combustion only dates back to 1970’s. In 1973 Sulzmann et al. [14] theoretically calculated the detection limits for combustion intermediates and products with line-center absorption and derivative spectroscopy using tunable diode lasers and hypothesized that detection limits
up to 3 to 4 orders of magnitude higher than conventional correlation spectroscopy was possible with TDLAS. Soon after, in 1977, Hanson et al. [11] used a tunable diode laser to capture high-resolution spectra of CO in the post flame region of a flat flame burner and determined fundamental spectroscopic parameters like line-strength and collisional line-width for the transition. In 1980, Varghese et al. [16] used TDLAS to scan 33 rotational-vibrational transitions between 4.44 - 4.78 \( \mu \text{m} \) of the CO molecule mixed in Ar and N\(_2\) to provide an improved value of the fundamental band strength of CO which maybe needed in connection with such applications in combustion. Cassidy et al. [17] applied a similar technique to detect trace gases (CO and CO\(_2\)) with a short-external-cavity InGaAsP diode laser using mode control. In this technique, a short cavity, external to the laser was used to force the laser to emit light in a single mode. The method was advantageous since it could use easily available multi-mode lasers and force them to operate in a single mode with significantly wide (~30 cm\(^{-1}\)) tunability range. In 1996, Nagali et al. [18] developed a diode laser system to monitor CH\(_4\) concentration in high temperature environments. A heated static cell was used for the purpose and fundamental spectroscopic parameters for transitions in the 1.66 \( \mu \text{m} \) range were determined in the 400 to 915 K range. In addition, a corrected third-order polynomial for the partition function was also constructed to approximate line-strengths in the intermediate temperatures.

The late 1990’s and early 2000’s saw a rapid growth in the development of diode laser systems for combustion flows mainly due to the easy availability of telecom grade tunable diode lasers. Mihalcea et al. [19] used a fast-flow multipass cell containing probe sampled combustion gases from a laminar, premixed methane-air flame to monitor CO, CO\(_2\) and CH\(_4\) concentration. An external cavity diode laser in the range of 1.49 to 1.58 \( \mu \text{m} \) and a DFB (distributed feedback) diode laser near 1.65 \( \mu \text{m} \) was multiplexed using optical fibers and absorption results were compared to absorption data from HITRAN 96 database [20]. Since most of the HITRAN database were tabulated at temperatures much lower than real combustion temperatures, significant deviations were found in the values of the fundamental spectroscopic parameters which were corrected. Similar experimental setup and laser system were used by same authors [21] to measure concentrations of NO\(_x\), CO, CO\(_2\) and O\(_2\) in combustion gases as well. Nelson et al. [22] in 1998 developed a TDLAS based spectrometer to measure air pollutant emission from on-road motor vehicles. The system made use of a novel setup using a complex arrangement
of beam splitters, mirrors and retro reflectors to simultaneously find the concentration of NO, CO and CO$_2$ from on-road vehicles. Upschulte et al. [23], in 1999, used a multisection InGaAsP diode laser that could be broadly tuned over almost 100 nm quasi-continuously only by the use of injection current. The laser was used to detect CO and CO$_2$ at room temperatures and CO, H$_2$O and OH in the exhaust of a CH$_4$ - air flame. Zhou et al. [24], in 2003, developed a single diode laser sensor capable of measuring in situ temperature and concentration of water vapor inside a flame. The authors found promising transitions of water vapor near the 1.8 $\mu$m region that were good candidates for TDLAS measurements. The study also outlined some of the major criteria for selection of spectroscopic parameters to design a temperature and concentration sensor using only a single laser diode. Some of the important criteria outlined are i) the transitions need to be close enough so that they can be accessed by a single DFB diode laser (which generally have maximum tunability of ~2 cm$^{-1}$) ii) they also need to be isolated from other nearby transitions and iii) have lower state energy values (E''') that are well separated. The last condition ensures that the sensor has a high sensitivity so that small errors in spectral absorbance measurements would not be magnified in temperature determination. Following similar guidelines, Liu et al. [25], designed a sensor to detect temperature and concentration of water vapor in an industrial gas turbine. The sensor was capable of measuring temperature in the 300 - 1000 K range to an accuracy of $\Delta T$~2K for an optimized line pair and to $\Delta T$~ 5K for an alternative line pair thereby proving the practical utility of TDL sensing in harsh industrial environments.

All the work mentioned above employed the direct-absorption method for determination of temperature and concentration. However, significant work has also been done with the WMS-2f TDLAS for highly sensitive and rapid measurement of temperature and concentration of species. The experimental strategy of harmonic detection to monitor weak signals had previously been applied to the studies of electron / magnetic resonance, Zeeman and Stark spectroscopy, acousto-optical spectroscopy and even in frequency modulation spectroscopy [26] (FMS, which is similar to WMS, but with modulation frequencies much larger than the absorption line-width). Reid and Labrie [27] performed wavelength-modulated TDLAS experiments to compare with theory of infrared absorption spectroscopy of gases and found excellent agreement. Phileppe et al. [28], used this method for simultaneous detection of temperature, pressure and velocity in shock-heated oxygen flows. The authors used a high-frequency
(10 MHz) wavelength modulation of a AlGaAs laser diode superimposed on a relatively low-frequency (10 kHz) repetitive scan to capture the second harmonic absorption data. Using a least-squares fit the data was fitted to theoretical second harmonic lineshapes (as in Fig. 2.5) and temperature and pressure were thus determined. Velocity of the flow was determined from the Doppler shift of the absorption line shape. Kluczynski et al. [15], used a Fourier domain description to clearly show how different Fourier components combine with line-shape functions to give rise to the final analytical function as well as the background signals. The analysis also clearly explained why only a laser with a non-linear intensity modulation will have background ”nf” signals even without the presence of any absorption. Liu et al. [29] developed a method to use WMS-2f spectroscopy for temperature and gas concentration detection using highly blended and pressure broadened absorption spectra and large modulation depths. The authors used a wavelength modulated near-IR laser and gathered absorption data from pressure broadened water vapor lines in the in the temperature range of 296-800K and pressures ranging up to 20 atm. Modulations depths as high 0.8 cm$^{-1}$ were used for the purpose. Simulations based on HITRAN 2000 [20] database numbers showed confirmation of the method’s capability to be used for rapid detection of temperature and gas concentrations in high pressure environments. Liu et al. [30], also used the two-transition ratio-thermometry method with WMS-2f for rapid detection of temperature in gases. The method was validated by using it to measure temperature of water vapor in a static hot-gas cell. However, instead of using the single laser diode, the researchers used a multiplexed fiber system to couple together outputs from a 1392 nm and 1343 nm diode for the purpose. Using a digital lock-in-amplifier, the 2f peak heights were filtered out from a modulated signal that had a 170 kHz sinusoid riding on a 1kHz saw-tooth. These 2f heights were used to ascertain temperature of the gas. In 2005, Zhou, et al. [31] developed a fast IR diode laser sensor that uses WMS-2f spectroscopy to measure temperature and concentration of water vapor. Using a systematic analysis of the HITRAN data set for H$_2$O line transitions between 1 to 2 µm, the authors selected a line pair that was close enough to be scanned by a single diode laser sensor. The sensor was validated inside a heated static gas cell and a temperature readout rate of 2 kHz was achieved while measuring temperature of an laboratory scale burner. A number of similar works [32, 33, 4] document the development and application of TDLAS with WMS-2f detection for in-situ measurement of temperature and / or concentration in flames.
Although laser flicker noise is one of the major sources of noise, it is definitely not the only source. Significant problems arise due to window fouling, beam walking (dancing of the laser beam spot due to changes in optical density along the laser path because of the presence of flame) or partial blocking of the beam by dust and dirt often encountered in harsh, dirty environments [30, 29, 34]. All these problems result in wavelength-independent attenuation of the laser intensity incident on the detector. Corrections for these are hard to quantify because of their stochastic nature. A major breakthrough in this area was the calibration free 1f-normalized 2f detection for WMS-2f. Li, et al. [5] demonstrated how the 2f peak height in the WMS-method can be normalized by the 1f output. Since these disturbances affect the overall incident intensity i.e. \(I_0\), which is a term that appears as a common factor in both the 2f and 1f expression, normalizing the 2f height with 1f effectively cancels out the effect of the overall laser intensity on the lineshape. The work also proposed and validated an improved model that takes into account the frequency modulation-intensity modulation phase shift and was applicable for large modulation depths.

1.3 Tomography

Tomography is the science behind the reconstruction of a spatial distribution from “line-of-sight” (LOS) integrated measurements also called “projections”. Its most common use is in the field of medical imaging - e.g. in a CAT scan where images of human body parts (like human head) are created using sets of projections sampled at small angular intervals. Therefore, high-quality images can be reconstructed using the large amount of directional information that is generally collected. Since TDLAS signals are LOS values, a set of such measurements spanning a spatial domain would therefore represent a set of “integral-equations” where the unknowns are absorption values at local points that intersect with the beam path. Ideally, such a set of equation would therefore be solvable provided enough information about the field is gathered from the measurements. Due to reasons like lack of optical access, high cost of implementation or just sheer engineering difficulty, a typical tomographic set up in the areas of physical sciences, in general, and combustion, in particular, would generally allow for only a few spatial sampling intervals unlike in medical imaging. A simplification in the form rotational symmetry of a steady field can allow for 1 dimensional (1D) tomography using Abel’s inversion [35] using only 1 set of parallel probing beams. Lack of such simplifying assumptions necessitates the use of inversion tech-
niques for 2 dimensional (2D) de-convolution. This class of tomographic solution falls in the category of “limited-view tomography” and throws significant challenges to produce satisfactory deconvolution results. Figure 1.4 shows a simplified schematic of a typical tomographic TDLAS setup.

1.4 Tomographic TDLAS: Motivation, Objectives and Approach

All examples of TDLAS applications mentioned in the previous section either assumed a uniform temperature and concentration distribution along the line-of-sight of the laser beam or that the variations due to non-uniformity were within a nominal acceptable limit which would not affect the final result. An application of line-integrated methods such as TDLAS to regions where quantities of interest vary significantly requires tomographic methods, which are used to deconvolute such data thus producing spatially resolved reconstructions [36, 37]. Depending on how the unknown distribution varies tomography can be of several kinds -1D, 2D or 3D. This work focuses only on 1D and 2D tomography. 1D tomography typically refer to distributions with an axial symmetry i.e. the distribution only varies in the radial direction. Such de-convolutions are popularly handled using different numerical implementations of the Abel inversion [35, 38, 39]. For 2D distributions several algorithms have been used. The classical method
is the filtered-backprojection method (FBP) used routinely in medical imaging [40, 41] but in physical sciences and engineering, where number of angular views are low, limited-view tomographic algorithms like algebraic reconstruction technique (ART) [42, 43] or Tikhonov regularized inversion [44, 45, 46] are often used.

One of the key criteria for the success of a tomographic scheme is to have a large amount of directional information, which in the case of TDLAS translates into a large number of LOS measurements from different directions. Unfortunately, as mentioned before, lack of optical access together with high cost of equipment, poses substantial problems in creating a tomographic measurement system that produces highly resolved directional information in a practical application. In recent years, limited-view-TDLAS tomography in particular, has shown promising results for hyper-spectral techniques [47, 9, 7, 8]. In this approach, the lack of directional information is compensated with additional spectral absorbance information from a large number (~100) of different spectroscopic transitions [7]. The temperature and concentration of the domain are then taken as free parameters and a non-linear optimization scheme (usually simulated annealing) is used to find a solution that minimizes the norm of the difference between the measured and calculated projections. Ma, et al. [7] used this method to successfully to reconstruct temperature and concentration of combustion zones in recent years. The setup used a custom made fiber Fabry Perot tunable filter laser (FFP-TFL) that could scan the water vapor lines between 1333-1377 nm. The laser resembled an external-cavity tunable diode laser but with parts that were completely made of fiber optics components while the tunable filter was used to tune the instantaneous wavelength. However, the length of the laser equipment was significantly larger (~17 m ) than a normal external cavity laser. Among other tomographic methods applied to flames, optical tomography methods have been applied to image chemiluminescence of flames [48], difference frequency generation combined with computer tomography has been used to image concentration of carbon monoxide in laminar flames [49] and even electrical capacitance/resistance and impedance tomography has been attempted for flame tomography [50, 51, 52].

In context of tomography of flames for temperature and concentration measurement using TDLAS, one work that deserves special mention is the study carried out by Villarreal, et al.[53] which is also the chief motivation for our work on one-dimensional tomographic TDLAS of a McKenna burner using
two-line thermometry. In this work, the authors reconstructed the temperature and concentration of CO₂ issuing from the combustion of a methane and air from a premixed laminar flat flame burner - a McKenna burner. The laser used in this case was a cryogenically cooled lead-salt Fabry-Perot laser with multimode output. The burner was mounted on a translation stage which helped it to move in and out of the laser-detector line of sight. A monochromator positioned after the flame acted as a tunable filter that allows records of spectral absorbance at different wavelengths. The laser output was scanned by using a sawtooth current ramp and around 200 scans were recorded at each location (~3 sec time) for averaging. Such records were taken at 20 different locations which acted as inputs for the Abel inversion. Each
scan, covering over 1000 transitions at each location, was digitized to create high-resolution transmission records for all the transitions. They were then spline smoothed and Abel-inverted to produce local values for the transmission spectrum. Subsequently, temperature and concentration of the flame at each point was calculated by taking them as free parameters while fitting the digital records to the CO\textsubscript{2} spectrum.

It should be noted that this study did not use the two-line thermometry for temperature and concentration deduction. The use of a broad-band diode laser helped in acquiring spectrum data over a large number of transitions and thus aided in achieving high degree of accuracy in deducing temperature and concentration. Using TDLAS to obtain temperature and concentration with just two lines and non-optimized modulation amplitudes therefore throws significant challenges if the absorbance data for either of the lines are corrupted with high amount of noise - sources of which can be laser noise (also known as relative intensity noise, RIN [13]), beam walking and other noise sources inevitable in an experimental setting. Additionally, Abel’s inversion formula that was used in the work above was based on equi-spaced parallel rays, which by the authors’ own admission, is well suited with flames where the gradients are more or less evenly distributed. In the McKenna flame, however, the gradients are concentrated near the very edge of the burner with the center being almost a flat top. Hence, we answer the following questions – i) whether coarse-grid equispaced arrangement of rays for sampling of the flame property distribution is optimal, ii) if non-uniformly spaced rays can be used to minimize reconstruction errors and iii) whether we can use simple equispaced rays and still achieve high resolution reconstruction irrespective of the presence of measurement noise. In our 1-dimensional tomography work, we investigate these questions systematically by analyzing the two sources of errors that corrupt Abel’s deconvolution - the regularization error (accuracy of reconstruction) and perturbation error (precision of reconstruction). Design-of-experiment theory is employed to answer the question of optimal ray spacings and model distributions are used to determine accuracy of existing deconvolution algorithms as well as new ones that were developed for this work, which use non-uniform ray spacings. Finally, the best possible option that preserves high degree of accuracy while minimizing perturbation errors was chosen for the deconvolution. This work also lays down the theoretical ground work for 2D tomography where no axi-symmetric assumptions are used. The results for 2D reconstructions were processed using 1D data extrapolated in 2 dimensions.
Traditional WMS-2f TDLAS techniques, theories of which are explained in chapter 2, implicitly assumes that the concentration of the investigated zone is known to within $\pm 10\%$ approximately and temperature is unknown [30, 2]. When concentration is not known and varies significantly more than the limits prescribed, amplitudes of modulation cannot be “optimized” and traditional WMS-2f temperature detection does not work well. Hence the broader objectives of this work are a) to develop a method that employs TDLAS for measurement of temperature and concentration in combustion zones with non-uniform distributions with minimum degree of tomographic artifacts and b) find a method to use WMS-2f TDLAS where modulation amplitudes cannot be optimized. In particular, we aim to reconstruct spatially resolved temperature and concentration distribution of water vapor in a laminar, premixed flat flame issuing from a McKenna burner which is a very well characterized burner and used for creating “top-hat”-like stationary temperature and concentration profiles. As a first step, 1D tomography or Abel’s axi-symmetric deconvolution is carried out to reconstruct the temperature and concentrations of the flame and then the 1D experimental data is extrapolated in 2D to reconstruct the same profile in 2 dimensions without the use of the axi-symmetric assumption.
Chapter 2
TDL Absorption Spectroscopy

2.1 Theory: Absorption Spectroscopy & TDLAS

Absorption spectroscopy aims at measuring the wavelength dependent energy absorbed by different molecules (e.g. oxygen, carbon-dioxide, water vapor etc. in atmosphere or flue gases after combustion) when they interact with electromagnetic radiation. From basic quantum mechanical results, we know that atoms and molecules can exist in only specific quantum states having a discrete amounts of energy. Furthermore, total energy of a molecule (which does not include kinetic energy) is stored in 3 different modes: rotational, vibrational and electronic or \( E_{\text{tot}} = E_{\text{rot}} + E_{\text{vib}} + E_{\text{elec}} \) [54]. Because of quantized internal energies, changes in quantum states leads to discrete changes in energies which correspond to energies of emitted or absorbed photons. This is the reason for the existence of selective wavelengths for emission or absorption – also called the emission or absorption spectra [54]. Using tunable lasers with well-characterized wavelengths, it is possible to measure the spectral energy absorbed by molecules, most commonly due to changes the rotational and / or vibrational states [24, 16, 55, 11].

2.1.1 Beer-Lambert’s Law

The fundamental theoretical principle of absorption spectroscopy is explained by the Beer-Lambert’s law (refer Fig.1.1) which relates the incoming and outgoing intensities of monochromatic light passing through an absorbing medium of uniform temperature and concentration [3]

\[
\left( \frac{I_t}{I_0} \right)_\nu = \exp(-k_\nu L) = \tau(\nu) \tag{2.1}
\]

where \( I_t \) and \( I_0 \) are transmitted and incident intensities, respectively, \( \nu \) is the instantaneous wavenumber, \( k_\nu L \) is the spectral absorbance (also sometimes represented as \( \alpha_\nu \)) and \( \tau(\nu) \) is the spectral transmission coefficient, \( L \) [cm] being the path length of absorption medium. The spectral absorption coefficient \( k_\nu [\text{cm}^{-1}] \) is represented as

\[
k_\nu = P \sum_{i=1}^{K} X_j \sum_{i=1}^{M_i} S_{i,j}(T) \phi_{ij}(\nu - \nu_{0,i}) \tag{2.2}
\]
where $P_{\text{atm}}$ is the pressure of the absorbing medium and $X_j$ is the mole fraction of the $j$-th absorbing species. $S_{i,j}[\text{cm}^{-2}\text{atm}^{-1}]$ is the line-strength (a fundamental spectroscopic parameter) corresponding to the $i$-th spectroscopic transition of the $j$-th species centered at $\nu_{0,i}$ and $\phi_{i,j} [\text{cm}]$ is the line-shape function from a mathematical model (refer to section 2.1.2) such that

$$\int \phi_{i,j}(\nu - \nu_{0,i})d\nu = 1 \quad (2.3)$$

Therefore, for optically thin transitions ($\alpha < 0.1$ and $1 - \exp(-\alpha) \approx \alpha$), Eq. 2.1, 2.2 and 2.3 can be combined as

$$\alpha_{\nu} = k_{\nu}L = LP_{\text{atm}} \sum_{j=1}^{K} X_j \sum_{i=1}^{M_i} S_{i,j}(T)\phi_{i,j}(\nu - \nu_{0,i}) \quad (2.4)$$

For most combustion applications it is reasonable to assume the total pressure $P$ as constant whereas temperature and concentration may vary along the line-of-sight (LOS). So, for a non-homogenous absorbing medium the above equation needs to be modified as follows:

$$\alpha_{\nu} = P \int_{0}^{L} \sum_{j=1}^{K} X_j(l) \sum_{i=1}^{M_i} S_{i,j}(T(l))\phi_{i,j}(\nu - \nu_{0,i})dl \quad (2.5)$$

### 2.1.2 Line-shape Functions

Lineshape functions (i.e. the quantity $\phi_i$) are analytical expressions that provide a measure of the breadth of an absorption transition or the line width, also known as full-width-at-half-maxima (FWHM, represented by $\Delta\nu$, refer Fig. 2.1). Simply put, this quantity determines how much the absorption changes as one moves away from the center wavelength ($\nu_0$, refer Fig. 2.1) of the transition. Such spectral line-shapes exist due to the phenomenon of line broadening [54]. Line-broadening mechanisms for a transition can be grouped into the following mechanisms: a) homogeneous broadening and b) inhomogeneous broadening. Homogeneous broadening mechanisms are mechanisms that affect all molecules in the same way while inhomogeneous broadening are mechanisms that affect different groups of molecules in different manners. The three major analytical line-shape functions that are discussed here are i) Lorentzian profile ii) Gaussian profile and the iii) the Voigt profile.
Natural Broadening. The major homogenous broadening mechanisms are natural lifetime broadening and collisional (pressure) broadening which give rise to Lorentzian lineshapes. The former stems directly from Heisenberg’s Uncertainty Principle which relates the uncertainty in the energy of the different states with their corresponding finite lifetimes. As per the Uncertainty Principle, the uncertainty in energy of the i-th level is

\[ \Delta E_i \geq \frac{\hbar}{2\pi \tau_i} \]  \hspace{1cm} (2.6)

If we assume the lifetime of the upper and the lower states to be \( \tau' \) and \( \tau'' \) respectively, and combine these uncertainties together, we get a range of energies over which the transition has a finite non-zero probability of being measured. This range is characterized by the full-width-at-half-maxima (FWHM) or the line-width. Using Planck’s relation of \( E = h\nu \), Eq. 2.6 for combined energy uncertain can be written as

\[ \Delta \nu_n = \frac{1}{2\pi} \left( \frac{1}{\tau'} + \frac{1}{\tau''} \right) \]  \hspace{1cm} (2.7)

The line-shape function can be derived by assuming the atomic system as a damped oscillator and the corresponding analytical expression is

\[ \phi_L(\nu) = \frac{1}{2\pi} \frac{\Delta \nu_n}{(\nu - \nu_0)^2 + (\frac{\Delta \nu_n}{2})^2} \]  \hspace{1cm} (2.8)

where \( \Delta \nu_n \) is the “natural” line-width of the transition and \( \nu_0 \) [cm\(^{-1}\)] is the line-center frequency.

Collisional Broadening. The other major homogenous broadening mechanism - the collisional broadening mechanism, arise due to the intra-molecular/atomic transfer of energy between different modes (rotational mode and / or vibrational mode etc.) that occur during collisions. Due to these collisions, the lifetime of a molecule or atom in a particular state becomes even shorter than the natural lifetime \( \tau_i \) as mentioned before. The shortening of lifetime leads to an increased \( \Delta \nu \) and, thus, a broader line-shape. The collisional broadening has the exact same expression as in equation 2.8, with \( \Delta \nu_n \) replaced by the collisional broadening width \( \Delta \nu_c \) where

\[ \Delta \nu_c = P \sum_j X_j (2\gamma_j) \]  \hspace{1cm} (2.9)
Here, $\Delta \nu_c$ is the collisional broadening in a mixture containing “j” species and $\gamma_j$ [cm$^{-1}$ atm$^{-1}$] is the coefficient for broadening for the species being probed colliding with species “j” in the mixture. The broadening coefficient varies with temperature of the mixture (T) through the following relationship

$$\gamma_j(T) = \gamma_j(T_0) \left( \frac{T_0}{T} \right)^{n_j}$$  \hspace{1cm} (2.10)

where $n_j$ is the coefficient of temperature dependence and $T_0$ being a standard temperature – usually 296K. Pressure broadening, like natural broadening, is a homogeneous broadening mechanism and therefore follows a Lorentzian profile i.e. Eq. 2.8. The total FWHM for homogenous broadening mechanisms is given by [54]

$$\Delta \nu = \Delta \nu_n + \Delta \nu_c$$  \hspace{1cm} (2.11)

However, in most cases the natural broadening mechanism can be neglected due to the long lifetimes of the energy levels and only pressure broadening is considered.

**Doppler Broadening.** The Gaussian or Doppler broadening is an inhomogeneous line broadening mechanism that contribute significantly to absorption line broadening phenomenon. If absorbing molecules have a velocity component in the direction of the laser beam, then the spectroscopic transition experiences a Doppler shift. The velocity of molecules in a sample of gas follows a Maxwellian distribution.
and hence this phenomenon affects different molecules differently since molecules with different classes of velocities will have different Doppler shifts. The velocity distribution function can be used to derive the analytical expression for the Gaussian lineshape and is given by

$$\phi_D(\nu) = \frac{2}{\Delta \nu_D} \left( \frac{\ln 2}{\pi} \right)^{1/2} \exp \left\{ -4 \ln 2 \left( \frac{\nu - \nu_0}{\Delta \nu_D} \right)^2 \right\}$$  \hspace{1cm} (2.12)

The Doppler FWHM $\Delta \nu_D$ is given by

$$\Delta \nu_D = \nu_0 \left( \frac{8kT \ln 2}{mc^2} \right)^{1/2} = \nu_0 (7.1623 \times 10^{-7}) \left( \frac{T}{M} \right)^{1/2}$$ \hspace{1cm} (2.13)

where $\nu_0$ is the line-center frequency [cm$^{-1}$], $T$ is the absolute temperature, $M$ is the molecular weight in grams/mole and “c” is the speed of light.

**Voigt Profile.** In most cases, instead of being either a purely collisional (Lorentzian) or a purely Doppler line-shape, the extent of actual line-broadening is a combined effect of the above two major mechanisms. In such a situation, the actual broadening for a given transition is given by the convolution of the Doppler and the collisional broadening functions. Such a function is also known as the Voigt line-shape function. Analytically the Voigt line-shape function can be represented as

$$\phi_V(\nu) = \int_{-\infty}^{+\infty} \phi_D(u) \phi_C(\nu - u) du$$ \hspace{1cm} (2.14)

where “u” is a dummy variable. The exact expression for the Voigt line-shape function can be found by substituting the expressions for the Doppler and the collisional line-shape functions in Eq. 2.14. However, the following non-dimensional groups are commonly used to simplify the algebra:

1) the Voigt parameter “$a$”

$$a = \frac{\sqrt{\ln 2} \Delta \nu_c}{\Delta \nu_D}$$ \hspace{1cm} (2.15)

The Voigt parameter is a scaled ratio of the collisional broadening half-width and Doppler broadening half-width. Therefore, it is a measure of which broadening phenomenon - the Doppler or the collisional - plays a major role. For a Voigt parameter $a > 10$, the effect of Doppler broadening can be assumed to be small and a purely Lorentzian lineshape maybe assumed.
2) The non-dimensional line-position “w” which signifies the distance from the line-center

\[ w = \frac{2\sqrt{\ln 2}(\nu - \nu_0)}{\nu_D} \]  

(2.16)

3) The line-center magnitude for the Doppler line-function

\[ \phi_D(\nu_0) = \frac{2}{\Delta \nu_D} \sqrt{\frac{\ln 2}{\pi}} \]  

(2.17)

4) The integral variable “y”

\[ y = \frac{2u\sqrt{\ln 2}}{\Delta \nu_D} \]  

(2.18)

With the above substitutions, Eq. 2.14 can be re-written as

\[ \phi_V(\nu) = \phi_D(\nu_0) \frac{a}{\pi} \int_{-\infty}^{+\infty} \frac{\exp(-y^2)dy}{a^2 + (w - y)^2} = \phi_D(\nu_0)V(a, w) \] 

(2.19)

Here, the function \( V(a, w) \) represents the integral and is also known as the Voigt function. There are many numerical approximations for this integral but this work utilizes the algorithm outlined by J. Humlíček [56].

**Boltzmann distribution and local thermodynamic equilibrium.** All the equations mentioned above implicitly assume that the molecules in the field are in “local thermodynamic equilibrium”. In other words, the basic assumption for the application of TDLAS techniques is that the time scales of the flow field are significantly larger compared to the time taken by the molecules to redistribute their energy into the different available energy-modes as per the Boltzmann’s distribution function. To explain this, we reconsider the absorbance equation, Eq. 2.5. Assuming a homogenous temperature and concentration field, only one isolated spectroscopic transition, and only one absorbing species, this can be re-written in a simplified manner as

\[ \alpha_V = \text{PXSL}(\nu_0, T)\phi_V L \]  

(2.20)

where the term \( S(\nu_0, T) \) is the temperature dependent line-strength value for a given spectral transition. This quantity can be accurately calculated for a given temperature if its value at a standard temperature
is known. This is done using the Boltzmann’s population fraction equations given by

\[
\frac{n_i}{n} = \frac{g_i \exp(-\varepsilon_i/kT)}{Q}
\]  

(2.21)

where

\[
Q = \sum_i g_i \exp(-\varepsilon_i/kT) = Q_{\text{rot}} Q_{\text{vib}} Q_{\text{elec}}
\]  

(2.22)

Here, \( n \) is the total number of molecules / atoms, \( n_i \) is number of molecules / atoms in the \( i \)th energy level, \( g_i \) is the degeneracy of the level (states that have the same energy \( \varepsilon_i \)) and \( Q \) is the partition function which is a specific energy weighted summation over all the available levels. The relationship between the line strength values at one temperature with those at another can then be written as [57, 20]

\[
S_{\nu 0}(T) = S_{\nu 0}(T_0) \frac{Q(T_0)}{Q(T)} \exp \left[ -\frac{\hbar c E''_{\nu 0}}{k} \left( \frac{1}{T} - \frac{1}{T_0} \right) \right] \frac{1 - \exp^{-\frac{\hbar c v_0}{kT}}}{1 - \exp^{-\frac{\hbar c v_0}{kT_0}}}
\]

(2.23)

where \( T_0 \) is the standard state temperature (generally 296 K), \( E''_{\nu 0} \) is the lower state energy value of the transition and \( Q(T) \) is the temperature dependent partition function of the molecule. In calculating “look-up-tables” for ratio and temperature, the line-strength equation, Eq. 2.23, along with parameter values from the HITRAN / HITEMP database were used for computation. Deviations from “equilibrium statistics” can occur in certain situations – e.g. when the population of molecules in the section of interest is not large enough or when time scales of fluctuations in the field are very small. One example of the former situation is a flow where the Knudsen number (\( \text{Kn} = \lambda / L = \frac{\text{mean free path}}{\text{characteristic length}} \)) is high (~1) i.e. rarified flows. Since this work records measurements that are downstream of the reaction zone (in the flue gas region) the only characteristics dimension here is the beam diameter (~0.5 mm). On the other hand, mean free path of the molecules are expected to be of the order of several hundred nanometers (for temperatures of 1700 K) resulting in a Knudsen number that puts the measurement section firmly in the continuum region. The latter can occur in the flow downstream of a strong shock-wave or during expansion through the nozzle of high speed wind tunnel. In such cases it is important to check the validity of these equations by comparing the characteristic times of the fluid flow with the characteristic times required by the molecules to readjust to the changing flow field via collisions. Although the readjustment
times for translation and rotation are small, vibration readjustments can take significantly longer [58]. A measure of the time taken for a population to return to, after being perturbed from, equilibrium is given by the relaxation time, \( \tau \). The value \( \tau \) is a function of temperature, pressure and the molecule itself and can be calculated as [58]

\[
\tau = \frac{K_1 T^{5/6} \exp(K_2 / T)^{1/3}}{P(1 - e^{-\Theta_v / T})}
\]  

(2.24)

where \( K_1 \) and \( K_2 \) are constants depending on the molecule, \( T \) is the temperature, \( P \) is the pressure and \( \Theta_v = h \nu / k \), \( \nu \) being the frequency of vibration of the molecule that has been modeled as a harmonic oscillator. When the relaxation time, \( \tau \), can be considered small compared to the flow times the above equilibrium equations hold. Using this it can be shown that from oxygen molecules (\( O_2 \)) at 1 atm. pressure and 2000 K, the vibrational time scales are of the order of 1e-6 secs [58]. The equation above uses gross assumptions [59] and as such is only accurate for order of magnitudes comparisons. However, Millikan et. al’s [59] experimental study provides a good reference point to evaluate these time scales for different temperature, pressures and compositions in a more accurate manner. Since our measurements have time scales of the order of 1e-2 secs the assumptions stated above hold.

**Spectroscopic Database (HITRAN/HITEMP).** The HITRAN / HITEMP [20, 57] database is a spectroscopic database that lists experimentally verified values of the spectroscopic parameters like line-strengths, broadening half-widths, coefficient of temperature variations for those widths etc. for different molecules. It also lists coefficients for a polynomial approximation of the partition function \( Q(T) \) for different ranges of temperatures. Since many of those molecules are also of interest to the combustion research community, in recent years the database has also been extensively used in this field [55, 60, 61, 62, 5, 63, 32]. Spectroscopic values of the parameters for a molecule at specified wavelengths and given temperature can be queried and found from the database. The above equations can then be used to simulate absorbance values of different gases with different pathlengths and compositions.

### 2.2 Direct Absorption (DA) Spectroscopy

A distributed feedback (DFB) laser is a class of semiconductor laser where the active region has the structure of a diffraction grating [64]. In these lasers the emission wavelength (as well as the intensity) can be changed rapidly by changing the injection current to the diode. Similar change in wavelength can
be achieved by changing the temperature of the diode but typically changes due to temperature variation are significantly slower. The wavelength tunability of these distributed feedback (DFB) diodes using only injection current, though quite narrow (~2 cm\(^{-1}\) or ~ 60 GHz in the telecom range), is still wide enough to scan across individual absorption lineshapes completely. Typically, at a constant injection current and constant temperature, a DFB laser has an emission line-width (band-width) of a few megahertz (the ones used in this work has a line-width of 2 MHz), while molecular absorption transitions have line-widths of the order of a few gigahertz (refer Fig. 2.2). In other words, the emission band of the lasers can be considered monochromatic w.r.t the spectral transitions bandwidths. (Note : Line-widths / band-widths, \(\Delta \nu\), though generally expressed in terms of spatial frequency i.e. wave-numbers \([\text{cm}^{-1}]\) can be equivalently expressed in terms of temporal frequency or hertz by the relation \(\Delta f_r = \Delta \nu \cdot c\) where \(\Delta f_r\) represents the line-width in hertz and \(c\) is the speed of light). Narrow band-width emission along with temperature and current tunability makes these lasers ideal for TDLAS measurements.

Direct absorption (DA) spectroscopy – the simplest form of TDLAS requires the laser wavelength (the black curve in Fig. 2.2 ) to be scanned across an absorption feature of interest (the red curve in the same figure) by changing the injection current of the diode often using a low frequency \((f \approx 0.5 \text{ to } 1 \text{ kHz})\) sawtooth or triangular waveform. The spectrally resolved transmitted laser intensity \(I_t(\nu)\) is simultaneously recorded using a photo-detector. Figure 2.3 is a simulation of this phenomenon and explains the
method graphically. In this figure, the output power of the diode (i.e. the incident intensity $I_0$) for a given injection current is plotted on the right-hand side y-axis, while its emission frequency (wavenumber) is plotted on the left-hand side y-axis (but not to scale). In general the emission wavelength of a diode increases (and hence the frequency decreases) with the increase in injection current, which explains the inverted left-hand y-axis of the figure. The black curve in the figure represents the transmitted intensity after absorption from the gas sample. In a real experiment the incident intensity is found by fitting a lower-order polynomial through the non-absorbing (though noisy) wings of the transmitted intensity curve. This fitted polynomial forms the baseline that indicates zero-absorption which is required for the DA measurements. Assuming only one absorbing species (i.e. dropping the subscript “j” for X) and isolation from nearby transitions the spectral absorbance in Eq. 2.4 can be re-written for the i-th transition as [3]

$$\alpha_{\nu,i} = \text{PLXS}_i \phi_i (\nu - \nu_{0i})$$  \hspace{1cm} (2.25)

The area under the spectral absorbance curve is also called the integrated absorbance (as indicated in Fig. 2.3) which using the help of Eq. 2.3 can be represented by

$$A_i = \int_{-\infty}^{\infty} \alpha_{\nu,i} d\nu = \text{PLXS}_i (T)$$  \hspace{1cm} (2.26)
Generally the area is found by fitting a Voigt function through the transmitted intensity curve $I_t$, finding the spectral absorbance by subtracting the baseline from it and integrating over the spectral region of interest. Equation 2.26 clearly shows that the integrated absorbance is a linear function of the concentration and a non-linear function of temperature through the line-strength term. Therefore, the ratio of the integrated absorbances of the two different absorption lines (i.e. ratio $R = A_2/A_1$) will only be a function of temperature which can be determined by some algebraic manipulation by the following equation [3]

$$T = \frac{\hbar c (E''_2 - E''_1)}{\ln(R) + \ln(S_2(T_0)/S_1(T_0)) + \frac{\hbar c (E''_2 - E''_1)}{T_0}}.$$ \hspace{1cm} (2.27)

Here, $E''_i$ denotes lower state energy values of the two transitions as tabulated in the HITRAN / HITEMP database and $T_0$ is the reference temperature - generally set to 296 K. Based on this temperature found from Eq. 2.27, the line strength $S_i$ can be calculated and the concentration of the gas sample can be determined as

$$X = \frac{A_i}{S_i PL}.$$ \hspace{1cm} (2.28)

The accuracy of the DA method is limited by the accuracy of the baseline and Voigt function fitting. Since the baseline is found by fitting the polynomial to the “non-absorbing” wings, it is paramount that a transition which is reasonably isolated from neighboring transitions is used. Also it must be noted that the method relies on accurately finding the area under the spectral absorbance curve. Hence a 1% error (say) in finding the baseline does not translate to 1% error in the area but much more due to the non-linear relationship. Hence this method requires strong transitions that are significantly isolated.

### 2.3 Wavelength Modulation Spectroscopy (WMS)

The low frequency scanning ramp in DA is superimposed with a high frequency ($f \approx 150$ to $200$ kHz) sinusoid in wavelength modulation spectroscopy. This creates a modulated absorption signal which can be decomposed into its components comprising of the fundamental modulation frequency and its higher harmonics. Mathematically, the instantaneous laser frequency (wavenumber) or frequency modulation (FM) can be written as

$$\nu(t) = \bar{\nu} + a \cos(\omega t).$$ \hspace{1cm} (2.29)
where $\bar{\nu}$ is the central laser wavelength (which is a function of the slow moving scanning ramp; here it is considered constant w.r.t the rapid modulation to simplify the algebra), $a$ is the amplitude of modulation of the laser frequency while $\omega = 2\pi f$ is the angular frequency of laser frequency modulation. This modulation is achieved by modulating the injection current which simultaneously produces an intensity modulation (IM) as well. The intensity modulation (which is slightly non-linearity w.r.t to the injection current [65, 5]) generally has a phase shift w.r.t to the frequency modulation [28, 15, 27] and is represented by

$$I_0(t) = \bar{I}_0(1 + i_0 \cos(\omega t + \psi_1) + i_2 \cos(2\omega t + \psi_2))$$

(2.30)

where $\bar{I}_0$ is the average laser intensity at $\bar{\nu}$, $i_0$ is the linear (1f) and $i_2$ is the non-linear (2f) amplitudes of the incident intensity normalized by $\bar{I}_0$, $\psi_1$ is the linear phase FM/IM phase-shift and $\psi_2$ is the non-linear FM/IM phase-shift.

The transmission coefficient $\tau(\nu) = \tau(\bar{\nu} + a \cos(\omega t))$, is represented as periodic even function in $\omega t$ and expanded as a Fourier cosine series:

$$\tau(\bar{\nu} + a \cos(\omega t)) = \sum_{k=0}^{\infty} H_k(\bar{\nu}, a) \cos(\omega t)$$

(2.31)
The coefficients (harmonics) $H_k(\bar{\nu}, a)$ is found as

$$H_0(\bar{\nu}, a) = \frac{1}{2\pi} \int_{-\pi}^{+\pi} \tau(\bar{\nu} + a \cos \theta) d\theta \quad (2.32)$$

$$H_k(\bar{\nu}, a) = \frac{1}{\pi} \int_{-\pi}^{+\pi} \tau(\bar{\nu} + a \cos k \theta) d\theta \quad (2.33)$$

where $\theta = \omega t$. However, from Eq. 2.1, $\tau(v) \approx 1 - \alpha(v) = 1 - \text{PXLS}_i \phi_i(v)$. Substituting this into Eq. 2.32 and 2.33 it can be seen that:

$$H_0(\bar{\nu}, a) = - \frac{S_i \text{PXL}}{2\pi} \int_{-\pi}^{+\pi} \phi_i(v) d\theta \quad (2.34)$$

and

$$H_k(\bar{\nu}, a) = - \frac{S_i \text{PXL}}{\pi} \int_{-\pi}^{+\pi} \phi_i(v) \cos k \theta d\theta \quad (2.35)$$

in particular,

$$H_2(\bar{\nu}, a) = - \frac{S_i \text{PXL}}{\pi} \int_{-\pi}^{+\pi} \phi_i \cos 2 \theta d\theta \quad (2.36)$$
The detector signal, given by

\[ G_I(t) = G_0(\nu(t)) \cdot \tau(\tilde{\nu} + a \cos \omega t) \]  

(2.37)
can be fed into a “lock-in-amplifier” (explained in section 5.2) and different harmonics of this signal can be recorded. The 2nd harmonic of this signal is analytically represented as

\[ S_{2f}(\tilde{\nu}) = \frac{G_0}{2} \left( H_2 + \frac{i_0}{2}(H_1 + H_3) \right) \]  

(2.38)

where \( G \) is the opto-electrical gain of the system, \( \psi_1 \) has been taken as \( \pi \) and \( i_2 \) has been assumed to negligible. At the line-center, \( \nu_0 \), the 1st and the 3rd Fourier components of the absorbance curve (i.e. \( H_1 \) and \( H_3 \)) are always zero (since they are odd functions, refer Fig. 2.5) while \( H_2 \) is maximum. Therefore, the detector signal is only proportional to the 2f peak-height at the line-center. It can also be seen that \( H_2 \), in turn, is proportional to the concentration from Eq. 2.36(provided the term inside the integral is insensitive to variation in concentration). In such a situation, the ratio of the second harmonic component of two different absorption transitions at the line-center will then only be a function of temperature and hence 2-line thermometry can be applied to find temperature if a “look-up-table” is evaluated before-hand that relates the ratios to temperatures.

The success of this temperature determination approach depends on the condition that the line-shape integral be fairly constant with the variations in concentration. However, it has been shown [29, 27] that if the modulation index \( m = \frac{2a}{\Delta \nu} \) is around 2.2, then the line-shape integral ratio is, in fact, nearly constant. In other words, near \( m = 2.2 \), the second-harmonic ratio will be a weak function of concentration but a strong function of temperature. Hence a range of values of the 2f-peak height ratios, simulated in advance over a temperature range of interest, can act as a “look-up-table” for determining temperature from an experiment. This idea is explained in Fig. 2.6. It can be seen that even for concentration variation of \( \pm 10\% \) around the mean concentration, the ratio of 2f-peak heights are very close to each other and does not introduce significant errors if any of the concentration lines are used to determine temperature.

Some of the significant problems that arise in any TDLAS methods are to do with disturbances from window-fouling of the beam, beam blockage due to dirt particles and beam-walking (flickering
movement of the beam over the photodetector sensor because of small perturbations in the flow field). These are intensity attenuation sources and therefore create significant errors in absorption readings. In case of WMS-2f though, all the harmonics, like the 2f signal in Eq. 2.38, scale linearly with the instantaneous mean intensity of the laser [5]. It can be shown that the 1f signal is given by

$$S_{1f} = 0.5 G_0 I_0 (1 - f(P, X, L, T))$$  \hspace{1cm} (2.39)$$

For optically thin samples, the function $f(P, X, L, T) \ll 1$ and hence

$$S_{1f} = \frac{1}{2} G_0 I_0$$  \hspace{1cm} (2.40)$$

Since the 1f signal for an optically thin sample scales only with the laser intensity and is unaffected by the absorption from the gas, it becomes an effective tool for normalizing all other higher harmonics thereby taking care of problems such as beam-walking or window fouling. Neglecting the non-linear component of intensity modulation and with $\psi_1 = \pi$, the 1f normalized 2f signal is given by

$$\frac{S_{2f}}{S_{1f}} = \frac{1}{I_0} \left( H_2 + \frac{i_0}{2} (H_1 + H_3) \right)$$  \hspace{1cm} (2.41)$$

Figure 2.6: Temperature determination in WMS-2f
Since $i_0$ is specific to the laser, once that value is ascertained during the laser characterization, measurements can be taken without any further calibration of the process.

2.4 Noise

Typically, diode laser intensities suffer from high amounts of noise [13, 66, 12, 26, 27]. Figure 2.7 shows a record of an absorption signal scanned across the transition near 1392 nm at room temperature with an open path length of 100 cm for a free-space TO-can laser without an optical isolator. It is obvious from the figure that without the use of any noise reduction method measurements may be highly erroneous. The most significant of the different noise sources that corrupt laser intensities is known as the relative intensity noise (also called flicker noise) often abbreviated a RIN. The power spectrum of RIN typically shows a “$1/f^\alpha$” frequency dependence where $f$ is the frequency and the exponent $\alpha$ generally varies from 0.5 to 1.6 [13]. This means that RIN has higher power at lower frequencies and much lower power higher frequencies. Other noise sources like shot noise and thermal noise are white noise sources. They are typically very small in value relative to RIN and determine the noise floor [26, 13]. The DA spectroscopy method scans across absorption features at repetition rates of ~ 1kHz which is fairly low. Not surprisingly, this method is easily corrupted by the RIN. Scanning at higher frequencies (~50 to 100 kHz) is problematic since DFB diode lasers typically show a drastic drop in scanning range as scanning frequency is increased. Hence the DA method requires significantly strong yet isolated transi-
tions for recording an appreciably pure absorption line-shape. Interference from neighboring transitions, availability of diodes at any such possible wavelengths, prices and often availability of accompanying hardware (fiber optics, optical components etc) can inhibit the selection of such strong lines.

2.5 Remarks

The WMS-2f method improves over the DA method on the following counts: i) the method only uses the second harmonic component which theoretically has a zero-baseline (shown in Fig. 2.5) and therefore inaccuracies due to baseline-fitting is avoided, ii) since WMS-2f only uses the 2f peak height, a 1% error due to noise at the 2f frequency will imply a 1% error in 2f-height and iii) RIN (or flicker noise) – which is the biggest contributor to noise, is vastly reduced at the 2f frequency (~400-500 kHz). This is primarily because of two reasons – i) flicker noise amplitude is roughly inversely proportional (~ 1/f) to frequency, therefore, at the 2f frequencies, the power of noise is vastly reduced and ii) this method uses a “lock-in-amplifier” (LIA) to extricate the harmonics (section 5.2). The LIA acts as bandpass filter with extremely narrow bandwidth thereby only allowing a very small range of frequencies around the harmonic frequency to pass through. Additionally, the output signal of the LIA is the sum of all the signals at that frequency and since at a given frequency, noise has randomly distributed phase, most of the noise cancel out. Although in principle, WMS-2f signal has a zero background, other signals like intensity-modulation effects (also known as residual amplitude modulation or RAM [5]) will create a non-zero background. These background signals (or RAM) can however be easily calculated beforehand and accounted for. It is important to note here that the traditional WMS-2f method implicitly assumes that the range of temperature, though unknown, does not vary so much that the modulation index “m” is significantly different from 2.2 and the concentration is known to within ±10% of the mean value. In our tomography work this assumption does not hold good since the concentration of water vapor in the flame is expected to vary from equilibrium concentrations near the center of the flame [53, 67] to almost zero near the edge. Relevant strategies for determining temperature and concentration when concentration is completely unknown have been discussed in chapter 5. For non-homogeneous fields, just like the DA method, the 2f signals are also a path integral of the individual 2f signals for each point on the beam path as long as the total absorbance is small enough to be linearized (i.e. as long as \(1 - \exp(-\int \alpha(x)dx) \approx \int \alpha(x)dx\) holds). Therefore, local 2f heights can be de-convoluted from path
integral values. Finally, it must be noted that the noise sources listed above are not the only sources of noise; inevitable variations in flame, reflections from optical components etc. also contribute to noise but are difficult to be modeled or mitigated using techniques like WMS-2f. These errors contribute towards a noisy set of projections which in turn produce erroneous and/or unphysical reconstructions or artifacts in final reconstructions that are difficult to be distinguished from the actual distributions in the absence of any a-priori knowledge. Errors or artifacts stemming from these sources need to be mitigated in different ways and such methods are discussed in the chapters to follow.
Chapter 3  
Tomography: Algorithms and Numerical Experiments

As mentioned previously, in case of inhomogeneous fields, the detector signal is a path integrated absorption signal for both DA and WMS-2f spectroscopy. As such, these signals lend themselves to be tomographically deconvoluted to produce local values. This chapter gives an overview of the different 1D and 2D tomographic algorithms that are used for deconvolution. For 1D tomography, the most popular existing algorithms are discussed briefly. The sources of errors in 1D tomography are analyzed in detail and strategies to mitigate these errors are also discussed and evaluated. For 2D tomography, this work lays down the theoretical groundwork in terms of number of views required, number of rays per view required and deconvolution algorithm most suited for limited view tomography.

3.1 1D Tomography

Tomographic reconstruction of some cylindrically symmetric spectral quantity distribution from axially symmetric LOS integrated projections is frequently encountered in engineering and quite often in combustion [68, 38, 53]. These fall under the category of 1D tomographic inversion which was solved analytically by N. H. Abel [35] in 1826. Unfortunately, the solution is of limited practical utility since it requires the knowledge of the projection values in an analytical form which in reality is rarely, if ever, the case. For Abel’s inversion, researchers therefore generally numerically approximate the solution by using a finite difference description of the problem in either the field variable domain or the projection domain. These techniques are explained in the following sections.

The projection value of a ray passing through a rotationally symmetric field distribution is equivalent to Abel’s integral equation, i.e.

\[ p(r) = 2 \int_r^\infty \frac{f(\tilde{r}) \tilde{r} }{ (\tilde{r}^2 - r^2)^{1/2} } d\tilde{r} \]  

(3.1)

where \( p(r) \) is the value of the projection at a radial distance \( r \) from the center and \( f(r) \) is the rotationally symmetric field distribution. In Equation 3.1, the projection values on the left hand side are known, whereas the field distribution in the integral has to be determined. Formally, this constitutes a Volterra
IFK (integral equation of the first kind), i.e. it is part of a class of moderately ill-posed inverse problems [69]. In this context, the projection in Eq. 3.1 smoothen sharp variations in the function $f(r)$, and thus reduces changes in gradients of projections $p(r)$. In any practical implementation, however, small perturbations in measurements from experimental noise are invariably introduced in projections. In the inverse process, these perturbations are magnified, which pose significant challenges in its solution [68, 38].

Unlike most integral equations of the first kind, an analytical solution to Eq. 3.1 exists in the form of the well known Abel inversion,

$$f(r) = -\frac{1}{\pi} \int_r^\infty \frac{p'(\tilde{r})}{(\tilde{r}^2 - r^2)^{1/2}} d\tilde{r}$$ (3.2)

where $p'(\tilde{r})$ represents the derivative of the projection [35]. In Equation 3.2, the unknown field distribution is given explicitly. However, an exact solution requires an analytical expression for $p'(\tilde{r})$, which limits direct implementation of Eq. 3.2 [38, 68]. Instead, finite difference approximations using discretized projection values have found wide-spread applications [38]. As mentioned before, small perturbations in measurement sets that are inevitable in a real experimental setting, may introduce large errors during the inversion thereby creating a highly oscillatory final solution. A very common solution is to manually smoothen the data set and use large ray-spacings [38, 68]. This, however, leads to introduction of errors that are subject to vary with the experimentalists’ judgement and low spatial resolution respectively. A relatively more objective approach is the Tikhonov regularized Abel’s inversion [68] which attempts to alleviate this problem by turning the Abel’s ill-posed problem into a series of well posed problem and will be discussed in the subsequent sections.

Modern numerical algorithms for Abel inversions are predominantly based on work by Dasch [38], who reviewed “onion-peeling” and “three-point” discretized implementations of Eq. 3.1 and Eq. 3.2. In the absence of measurement noise, the only kind of error present in a reconstructed solution is the regularization error which constitutes a Taylor series expansion error [38]. This stems from the fact that the terms $-f(\tilde{r})$ and $p'(\tilde{r})$, of the analytical system of Eq. 3.1 and 3.2 are approximated by their finite difference descriptions using Taylor series expansion and converted into a linear algebraic system thereby introducing truncation errors. The magnitude of this error is therefore dictated by the grid spacing, finite
difference scheme and the function being reconstructed and as such is independent of variations in the measured data set.

While it is not a prerequisite, Dasch’s algorithms use uniform ray spacing, which has been maintained in subsequent research [70, 53, 68]. Research on loosely related multiangle elastic light scattering (MAELS) experiments, however, has shown that sampling of an experimental space at equal intervals does not always guarantee the best method [71, 44]. Using design-of-experiment theory [72], Burr et al. [44] successfully determined optimized angles that maximize the information content in MAELS measurements while mitigating a second kind of error - the perturbation error [44].
Here we focus on error mitigation where two deconvolution algorithms are considered. In each case, the implementation is generalized for arbitrary ray spacing. We then proceed to apply the design of experiment theory [72] to find if there exists an optimal ray arrangement that maximizes the information content in measured projections thereby minimizing the ill-conditioning of the Abel problem and investigate its effect on the precision and accuracy (perturbation error and regularization error) of the reconstructions in the presence of random measurement noise.

3.1.1 Abel’s Inversion: Projection and Deconvolution

For numerical implementations of Abel inversion, a rotationally symmetric domain is subdivided into annular rings with rays passing through the center of each annulus as illustrated in Figure 3.1. Thus, rays \( r_0, r_1, ..., r_{N-1} \) pass through annuli with labels \( a_0, a_1, ..., a_{N-1} \), respectively. With this definition of rays and annuli in place, the projection of field variables in Equation 3.1 is rewritten as a sum of integrals over the whole domain as

\[
p(r_m) = 2 \sum_{n=m}^{N-1} \int_{b_n}^{a_n} f(\tilde{r}) \frac{\tilde{r}}{(\tilde{r}^2 - r_m^2)^{1/2}} d\tilde{r}
\]

where the lower integration limit is

\[
b_n = \begin{cases} 
  r_n, & n = m \\
  a_{n-1}, & n > m 
\end{cases}
\]

If the field variable \( f(r) \) is approximated by a Taylor series expansion around \( r_i \), all integrals can be precalculated. Thus, the system of analytical equations (Eq. 3.3a) is represented by the matrix equation

\[
Af = p
\]

where the vectors \( f = [f_0, f_1, \ldots f_{N-1}]^T \) and \( p = [p_0, p_1, \ldots p_{N-1}]^T \) contain discretized field variable values and projection values, respectively. Furthermore, the \( N \times N \) matrix \( A \) acts as the “projection matrix” where elements \( A_{mn} \) define the weight of the \( n \)-th annulus contribution to the projection value of the \( m \)-th ray.
In a similar fashion, the deconvolution of the projection values in Equation 3.2 can be rewritten as

$$f(r_m) = -\frac{1}{\pi} \sum_{n=m}^{N-1} \int a_n \frac{p'(\tilde{r})}{b_n (r^2 - r_m^2)^{1/2}} d\tilde{r} \quad (3.4a)$$

In this case, the derivative of the projection measurements $p'(r)$ is substituted by their Taylor series expansion around $r_1$. Precalculating integrals Eq. 3.4a yields the matrix equation

$$f = Dp$$

(3.4b)

where the $N \times N$ matrix $D$ represents the “deconvolution matrix”. Ideally, an exact inversion would yield $D = A^{-1}$, which, however, does not hold in practical algorithms due to limitations of finite difference schemes used in the Taylor expansions.

### 3.1.2 Algorithms

Numerical algorithms for Abel inversion are either based on projection or deconvolution, which correspond to Eqs. 3.3a/3.3b and 3.4a/3.4b, respectively. While conventional algorithms assume uniform ray spacing, in this work we re derive the projection and/or deconvolution matrices for arbitrary ray spacing. Outlines of the algorithms are given below, whereas detailed derivations are given in the appendices.

- **Onion Peeling (OP)**

  The onion peeling algorithm works by constructing the projection matrix $A_{OP}$ using Eq. 3.3a. Inside the integral, function values $f(r)$ are considered constant inside each annulus. For OP, integration along a ray yields

  $$p = A_{OP}f + \delta p$$

  (3.5)

  where

  $$\delta p = \delta A_{OP}f^*$$

  (3.6)

  is an error estimate that scales with the first derivative of the field distribution.
• **Abel Three Point (ATP)**

The classical ATP algorithm approximates the deconvolution matrix $D_{ATP}$ numerically starting with Eq. 3.4a. In the integration, the derivative of the projection is developed in a two-term Taylor series expansion, where $p'(r_i)$ and $p''(r_i)$ are subsequently approximated by central difference schemes. The resulting scheme produces

$$f - \delta f = D_{ATP} p$$

where

$$\delta f = \delta D_{ATP} p'''$$

Here the discretization error is proportional to the third derivative of the projection $p'''$. This scheme assumes that at the center, $p(r_{-j}) = p(r_j)$ effectively making the first derivative of the projection equal to zero when $r = 0$. This is reasonable since, for field variables with bounded values, the projection is expected to be zero at $r = 0$. Additionally, in order to be consistent with Dasch’s implementation [38], the derivatives at the edge are computed by assuming a ghost annulus outside of the domain.

• **Three-Point Projection (TPP)**

Similar to OP, the TPP algorithm uses Eq. 3.3a to construct the projection matrix $A_{TPP}$. For the integration, the field variable $f(r)$ is approximated by a Taylor series expansion where the first two and three terms are retained for two variants TPPa and TPPb respectively. Away from the boundaries, $f'(r_n)$ and $f''(r_n)$ are approximated by three-point central differences, resulting in the two schemes

$$p = A_{TPP_a} f + \delta p_a$$

$$p = A_{TPP_b} f + \delta p_b$$

where

$$\delta p_a = \delta A_{TPP_a} f''$$

and

$$\delta p_b = \delta A_{TPP_b} f'''$$
Here the error estimates scale with $f''\star$ and $f'''\star$, respectively. At the boundaries, forward and backward difference are used at center and perimeter, respectively. While alternative implementations with ghost-points were investigated, the overall performance was comparable and the simplest implementation was retained.

### 3.1.3 Errors

Errors in an Abel inverted solution can be of two kinds - regularization error and perturbation error. In a practical measurement, projection values $p_{\text{meas}}$ are obtained as

$$p_{\text{meas}} = p_{\text{ex}} + \delta p_{\text{noise}}$$

(3.12)

where $p_{\text{ex}}$ is the noise-less projection of the field variable according to the underlying physical measurement principle, whereas $\delta p_{\text{noise}}$ is the noise introduced by the measurement system, i.e. excitation and data acquisition.

In the following, perturbation error is interpreted as $\delta f_{\text{pert}} = A^{-1}\delta p_{\text{noise}}$, whereas regularization error $\delta f_{\text{reg}}$ is defined as the deviation of a reconstructed field distribution $f_{\text{rec}}$ from the actual distribution $f_{\text{ex}}$.

It is noted that in a practical measurement system, regularization error and perturbation error correspond to accuracy “$\varepsilon$” and precision “$\sigma$”, respectively.

- **Regularization Error**

The exact projection $p_{\text{ex}}$, is equivalent to the hypothetical relation

$$p_{\text{ex}} = A_{\text{ex}}f_{\text{ex}}$$

(3.13)

where $f_{\text{ex}}$ is the field variable sampled at $r_n$, and $A_{\text{ex}}$ is interpreted as a projection matrix where the integrals in Eq. 3.3a are exact. Any practical implementation of a projection matrix $A$ based on a Taylor expansion of order $K - 1$ will, however, introduce a discretization error, i.e.

$$A_{\text{ex}}f_{\text{ex}} = Af_{\text{ex}} + \delta A f^{(K)}\star = p_{\text{rec}} + \delta p_{\text{reg}}$$
where the last term corresponds to the regularization error introduced by the discretization process. Multiplying the deconvolution matrix $D = A^{-1}$ from the left yields

$$f_{\text{rec}} = f_{\text{ex}} + A^{-1} \delta p_{\text{reg}} = f_{\text{ex}} + \delta f_{\text{reg}}$$

where $f_{\text{rec}} \equiv Dp_{\text{ex}}$ is the reconstructed field variable distribution. Thus, the regularization error in the reconstructed field for an algorithm based on a projection matrix becomes

$$A^{-1} \delta p_{\text{reg}} = \delta f_{\text{reg}} = A^{-1} A A^{-1} p^{(K)*}$$ (3.14a)

where $A^{-1} p^{(K)*} \approx f^{(K)*}$ was used. Similarly, the regularization error for an algorithm based on a deconvolution matrix is obtained as

$$\delta f_{\text{reg}} = \delta D p^{(K+1)*}$$ (3.14b)

Therefore, we define a “regularization coefficient” as

$$\varepsilon_c = A^{-1} \delta A A^{-1}$$ (3.15)

Due to its definition, it is clear that the regularization error is proportional to derivatives of the field variable distribution. At the same time, both $\delta A$ and $\delta D$ are proportional to the discretization step.

In the following, discretization errors are estimated assuming derivatives of order unity. Figure 3.3 illustrates the regularization error as a function of grid point number and algorithm.

- **Perturbation Error**

As mentioned before, the perturbation error is $\delta f_{\text{pert}} = A^{-1} \delta p_{\text{noise}}$ (alternatively, precision of reconstruction - $\sigma$). While regularization error is difficult to be quantified a-priori, perturbation error is only a function of the noise level in measurement and geometry of the grid that spans the domain [38]. The amplification of perturbation error in the reconstructed solution is an area of concern in Abel’s inversion which stems from the inherent ill-posedness of the problem [68]. Hadamard described a mathematically well-posed problem as one that has the following properties - i) it must have a solution, ii) the solution must be unique and iii) the solution needs to be a continuous function of its inputs. Abel’s equation is
only mildly ill-posed because it does not satisfy the third criterion. The fact that it satisfies the first criterion is intuitive in the sense that the measured projections themselves are the manifestations of existence of the field. The second criterion is satisfied because one can see that as \( r \) approaches \( R \), the value of \( f(r) \) can be positively inferred from \( p(r) \) and further values or \( f(r) \) for \( r < R \) can be continuously calculated using the onion-peeling analogy [68]. However, the solution to the problem suffers from the fact that small measurement noise is amplified in the reconstructed field. This property of Abel’s problem has been discussed in literature extensively [68, 38] and only re-explained here briefly to provide context. Summarily, in case of solving Eq. 3.1 by onion-peeling, increasing the resolution of measurement by increasing the number of rays creates an \( A \) matrix with rows very similar to each other till a point is reached where the matrix becomes rank deficient. Added to this is the fact that reconstruction errors progressively increase towards the center of the domain since the reconstruction errors at a smaller radii are contaminated by the errors at larger radii due the ‘onion-peeling’ method of solution.

Solution to Eq. 3.2 requires estimation of the local derivatives of projections by use of finite difference methods. Increasing the resolution of the grid increases the noise amplification while calculating local derivatives since the spatial distance between points decreases but noise levels remains unaffected. This is due to the fact that noise in measurements at different locations are independent and uncorrelated. Quantitatively speaking, an estimate of the magnification of measurement noise in the solution can be obtained by using perturbation theory [68] with Eq. 3.4b and it can be shown that

\[
\frac{||\delta f||}{||f||} \leq \text{cond}(A) \frac{||\delta p||}{||p||}
\]  

Eq. 3.16 shows that higher the condition number of the \( A \) matrix, higher the maximum level of error \((||\delta f||/||f||)\) in solution due to the noise level in projections \((||\delta p||/||p||))\). A small condition number (close to unity) implies a well-posed problem, in the sense that small variations in measurement sets remain small in the final solution, while a large condition number would imply an ill-conditioned problem where similar smaller variations are magnified in the solution. The condition number of a matrix can be calculated by taking the ratio of the maximum to the minimum singular values of the matrix i.e. \( \text{cond}(A) = \omega_{\text{max}} / \omega_{\text{min}} \) where \( \omega_j \) denotes the singular values of \( A \). The singular values in turn can be
Figure 3.2: Comparison of singular values in the onion-peeling and ATP algorithms using 15, 30 and 50 rays found by performing a singular value decomposition of $A$ i.e. expressing the matrix as

$$A = U \Sigma V^T$$

with $U, V$ being the two orthogonal matrices that form an orthonormal basis for the data space and state space while $\Sigma$ is the diagonal matrix containing the singular values $\omega$ which are non-negative and arranged in a decreasing order. Using the property that the inverse of a orthonormal matrix equals its transpose, one can write

$$f_{\text{rec}} = \sum_{j=1}^{N} \frac{u_j p_{\text{meas}}}{\omega_j} v_j = \sum_{j=1}^{N} \frac{u_j p_{\text{exact}}}{\omega_j} v_j + \sum_{j=1}^{N} \frac{u_j \delta p_{\text{noise}}}{\omega_j} v_j$$

where $f_{\text{rec}}$ is the noisy reconstruction of the function, and $p_{\text{meas}} = p_{\text{exact}} + \delta p_{\text{noise}}$, denotes the noisy projections. Figure 3.2 shows the singular value spectrum of the Onion-peeling and the Abel’s 3pt. algorithm. One can see that the singular values of both span several orders of magnitude and with increasing number of rays the singular values progressively decay towards zero. Therefore, to have a bounded solution, $u_j p_{\text{exact}}$ should approach zero at a faster rate rate than $\omega$, which is generally satisfied for unperturbed data. However, in the presence of noise, this is not the case and the second term in Eq.
Figure 3.3: Regularization coefficient variation with rays

3.18 shows how the smaller singular values amplify the noise in the solution till it overwhelms the actual solution as one increases the number of rays. This is also the reason, why experimentalists commonly use sparse grids (small number of rays) [38] in the case of onion-peeling to suppress noise in the solution. 

Fig. 3.2 also shows the reason for the superior noise reduction capability of ATP. algorithm as compared to onion-peeling: for the same number of rays, the 3pt. algorithm has a higher singular value spectrum than onion-peeling and therefore less prone to noise amplification.

- **Error Metrics**

For regularization error, the $||L||_2$ norm of the deviation vector $(f_{\text{exact}} - f_{\text{rec,noiseless}})$ is used as a metric (alternatively, accuracy of reconstruction - “$\varepsilon$”) in this work. Here $f_{\text{exact}}$ is the exact analytical distribution under consideration while, $f_{\text{rec,noiseless}}$ is the set of reconstructions from projections sampled from the set of exact “simulated projection measurements” that is not corrupted by noise. For perturbation error, the $||L||_2$ norm of the deviation vector $(f_{\text{rec}} - f_{\text{rec,noiseless}})$ has been used as metric (alternatively, precision of reconstruction - “$\sigma$”) in this work. Here $f_{\text{rec}}$ is the reconstruction of the field using projections sampled from the set “simulated projection measurements” that is corrupted with noise.

- **Merit of Algorithms**

Before we start analyzing the effects of different error mitigation strategies on the accuracy and precision of individual reconstructions, it is worthwhile to analyze the effects of the 3 algorithms - OP, ATP and
Figure 3.4: Local noise and regularization coefficient variation with number of rays

versions of TPP on reconstructions without reference to any particular distribution. This will help in determining the best algorithm among the three and can therefore be further utilized for this work.

With regards to perturbation error, the noise coefficient as used by Dasch [38] is an useful metric to judge algorithms. The error in reconstruction, for equi-spaced rays, due to measurement noise can be written as

\[ \delta f_i = \alpha \sqrt{\frac{1}{N-1} \sum_{j=0}^{N-1} D_{ij}^2} \]  

(3.19)

here \( D_{ij} = D_{ij}/\Delta r \), \( \Delta r \) being the distance between adjacent rays, \( \alpha \) being the rms fluctuation of each projection (this formula assumes that rms fluctuations of each projection is uncorrelated, independent of the field, and equal to each other) and \( D_{ij} \) being the elements of the deconvolution matrix \( D \) as defined by Dasch (also in appendix). The local noise coefficient is given by

\[ \sigma_c = \sqrt{\frac{1}{N-1} \sum_{j=0}^{N-1} D_{ij}^2} \]  

(3.20)

Figure 3.3 shows the radial variation regularization coefficient with variation in number of beams. Since a higher value of \( \varepsilon_c \) indicates a deteriorating level of accuracy, it can be seen that the accuracy of the solution decreases towards the center of the domain. This is expected since the Abel’s solution solves
the problem from outside to inside and hence all the errors incurred on the outside of the domain cumulatively add up towards the center. For the same reason, the noise coefficient as well deteriorates towards the center (Fig. 3.4). Comparing between algorithms, it can be seen that increasing grid resolution by increasing number of rays helps for all algorithms other than OP. For OP, high number of beams hardly reduces the regularization coefficient significantly while it does increase the noise coefficient drastically. From the point of view of accuracy, it can be seen that TPPb is the best followed by ATP, TPPa and OP. As far as noise coefficient is concerned, increasing number of rays increases the noise coefficient drastically (therefore decreases precision). However, it must be noted that for the same number of rays, all the algorithms except ATP has similar noise coefficients. ATP visibly has superior noise suppression capabilities than others.

In order to gain a comprehensive comparative view of the performance of these algorithms we calculate the “rms” values of the noise and regularization coefficients as outlined in “Error Metrics” in section 3.1.3. Plotting the RMS value of regularization coefficient against the RMS value of noise coefficient gives us a strong basis to compare the different algorithms (this assumes that for the regularization error, the higher derivatives are of the same order and for the perturbation error the local uncertainties are of similar orders). Figure 3.5 shows that for any given level of regularization error, the ATP algorithm is
least prone to perturbation error.

3.1.4 Strategies for Error Mitigation

Regularization error or accuracy of reconstruction (\( \varepsilon \)), is a direct function of the grid spacing, order of the finite difference scheme employed and the higher derivatives of the function itself. Intuitively, the accuracy can be increased by increasing either the number of rays or the order of the scheme or both. However, employing increased number of rays will also increase the condition number of the projection matrix. This effect is shown for all the algorithms in Fig. 3.6. As explained before (refer Eq. 3.16), a higher condition number has the tendency of amplifying any oscillation present in the data, thereby making the solution more susceptible to measurement noise. Hence, the subsequent discussions deal with mitigation of perturbation error with a fixed number of equi-spaced rays.

- **Tikhonov Regularization**

Destabilization of reconstructed solution due to the magnification of measurement noise can be mitigated by using regularization techniques [73]. Daun et al. [68] demonstrated the technique by reconstructing normalized soot-volume fraction data in a circular symmetric domain. In short, the system of equations is augmented by using an additional set of N-equations : \( \lambda Lf = 0 \) where \( L \) equals the discrete version of
the $\nabla$ operator, i.e.

$$
\mathbf{L} = \begin{bmatrix}
1 & -1 & 0 & \cdots & 0 \\
0 & 1 & -1 & \cdots & 0 \\
0 & 0 & \ddots & \ddots & \ddots \\
& & & & -1 \\
0 & 0 & \cdots & 0 & 1
\end{bmatrix}
$$

(3.21)

and $\lambda$ is a parameter that controls the extent of regularization applied. A solution to this overdetermined system is sought as

$$
\mathbf{f}_\lambda = \arg\min \left\{ \left\| \mathbf{A} \mathbf{f} - \mathbf{p} \right\| \right\}
$$

(3.22)

It is easily seen that the presence of $\mathbf{L}$ promotes a smoother solution by limiting the derivative values of field variable. The regularization parameter lambda controls the relative weightage placed on accuracy of the solution and smoothness of the solution during the solution search. A smaller $\lambda$ implies a solution which is highly oscillatory but one that explains the measured projections (including the noise) better than one with a higher $\lambda$ which will have a smoother solution but have a higher deviation from the measured solution. Obviously, the proper choice of $\lambda$ is critical for a high quality solution. Different methods exist for choosing the proper regularization parameter - e.g. the discrepancy principle, the L-curve criteria or the generalized cross-validation [74, 75, 76, 73]. In this work we follow the “L-curve” criterion [73]. The method which has been explained in literature [73, 68, 77] is briefly re-explained here. The L-shaped curve (Fig. 3.7) can be obtained by plotting the norm of the solution $||\mathbf{F}||$ versus the norm of the residual vector $||\mathbf{AF} - \mathbf{P}||$ for different levels of regularization i.e. for different values of $\lambda$. The upper left part of the plot points to values of $\mathbf{F}$ that accurately solve the linear equation $\mathbf{AF} = \mathbf{P}$ but are also highly oscillatory because of amplification of the noise in the data, whereas the lower right portion of the curve represent the smooth but over regularized solutions and hence not very accurate. The ideal value of the parameter hence lies somewhere on the downward sloping section of the curve. Fig. 3.7 shows such an L-curve for a representative axi-symmetric distribution reconstructed from synthetic noisy data using 25 rays. Manual investigation reveals that the optimal value of the regularization parameter lie somewhere between 0.01 and 1. While, there are a variety of methods to choose the proper regularization parameter
we choose three values of lambda $\lambda = 0.063, 0.1$ and $0.3$ using the 'L-curve criterion' [73] as a guideline for the subsequent numerical experiments.

- **Averaging**

In a real experimental situation the most frequent method of mitigation of perturbation noise is sample averaging. The advantage of averaging over Tikhonov regularization is that in Tikhonov regularization over-regularized solutions while suppressing noise, will also affect the actual distribution. However, since

$$p_{\text{meas}} = p_{\text{ex}} + \delta p_{\text{noise}}$$  \hspace{1cm} (3.23)

it is obvious that for sufficiently high number of samples, $p_{\text{meas}} \to p_{\text{ex}}$, assuming that $\delta p_{\text{noise}}$ has a normal distribution. If $p_{\text{meas}}$ is very close to $p_{\text{ex}}$ then it is obvious that the regularization required by Tikhonov will be significantly lower even for high-resolution beam spacings and hence will preserve both the accuracy and precision of the distribution better.

### 3.1.5 Design-of-Experiments (DOE)

As discussed before, the condition number of the matrix $A$, provides a means to quantify the maximum extent of noise amplification possible in the final solution. Since, the arrangement of rays solely determines the value of $A$ and hence the condition number, it is worthwhile to investigate the idea if there exists an optimal beam arrangement that is least prone to measurement noise amplification for a given
number of rays. The intuitive way of achieving it would be to look for an arrangement that minimizes the condition number of the \( A \) matrix directly (also known as E-optimality [72, 44]). However, the condition number, which is nothing but the ratio of the biggest and the smallest singular values obtained from the singular value decomposition of the \( A \) matrix, does not take into account the parameter co-variance. Therefore, a more formal method of optimization in this case, would be the D-optimality [72, 44] which takes into account the parameter co-variance. A demonstration of the method is documented at length in Burr, et al.’s [44] work on optimization of measurement angles for the measurement of soot volume fraction and is hence briefly touched upon here. Summarily, if it is assumed that the measurements in the vector \( p \) are mutually independent and belongs to an unbiased normal distribution with standard deviation \( \alpha \), then the quantity 
\[
\chi^2(f) = \sum_{i=1}^{N} \left[ \frac{a_i f - p_i}{\alpha_i} \right]^2
\]  
(3.24)
is known as the chi-squared function (here, \( a_i \) is the i-th row of the matrix \( A \)). Minimizing the chi-squared function is equivalent to maximizing the likelihood of
\[
p(p|f) = \prod_{i=1}^{N} \left( \frac{1}{\alpha_i \sqrt{2\pi}} \right) \exp \left( -\frac{(a_i f - p_i)^2}{2\alpha_i^2} \right)
\]  
(3.25)
and therefore, \( \chi^2(f^*) \) would indicate the extent to which the least-squared solution \( f^* = \text{argmin} \left[ \chi^2(f) \right] \), explains the measured data \( p \). If we scale the distribution width for each projection (i, being the projection or ray number) so that they are equal, then Eq. 3.24 can be re-written as
\[
\chi^2(f) = \frac{1}{\alpha^2} (Af - p)^T (Af - p)
\]  
(3.26)
and using the substitution \( f = f^* + \delta f \), Eq. 3.26 can be simplified to
\[
\Delta\chi^2(\delta f) = \chi^2(f^* + \delta f) - \chi^2(f) = \frac{1}{\alpha^2} \delta f^T A^T A \delta f
\]  
(3.27)
The quantity \( \Delta\chi^2(\delta f) \) therefore signifies a confidence interval traced out by \( \delta f \) sitting on the tail of the vector \( f^* \) for a given value of \( \Delta\chi^2 \) corresponding to a tabulated probability. This quantity is a hyper-ellipse in n-space and its volume is a measure of the how much the measurement noise in the projections
is amplified in the deconvoluted solution. Since the standard deviation and the $\Delta \chi^2$ values are constant, the only way to reduce $\delta f$ is to maximize the determinant of $A^T A$ which can be achieved by minimizing the objective function [72, 44]

$$S = \frac{1}{\det [A^T A]}$$

(3.28)

In a geometric sense, the $\Delta \chi^2=1$ surface is a hyper-ellipse in “n-dimensions” where “n” is number of rays or annuli. Its principal axes are aligned along the direction given by the elements of the column vector $V$ and scaled by the inverse of the singular values $\Sigma$. Therefore, making the singular values as large as possible minimizes the volume of the hyper-ellipse. Following this, the annulus edge location (and therefore the corresponding ray locations) were optimized to find the minimum of the objective function. Since this is a case of non-linear multivariate optimization, we chose simulated annealing to find the optimized distribution of annuli and rays. In order to achieve this a bound-constrained minimization process was used: $a_0$ was held constant at $r=0$, $a_n$ was held constant at $r = 1$ while all the other annuli edge positions were allowed to vary without having the same position as another. The last condition was achieved by checking the rank of the $A$ matrix and discarding those options that would result in a rank deficient matrix. The effect of change of ray positions on accuracy is analyzed using reconstructions of model (test) distributions.

3.2 Numerical Experiments

3.2.1 Test functions for accuracy of DOE and Tikhonov schemes

In order to investigate accuracy of a reconstruction algorithm it is important to use functions such that $f_{\text{exact}}$ and $p_{\text{exact}}$ are known in their analytical forms. Additionally, since optimization affects ray spacing of the domain it is expected that accuracy will be simultaneously affected by the grid spacing at different radial distances. Hence it is prudent to use functions that are locally similar to each other but shift across the domain radially. In light of such requirements, the performance of the algorithms were tested using the BASEX [79] set of basis functions proposed by Dribinski et al. The BASEX set of basis functions is mathematically represented as [79]

$$\rho_k(r) = \left(\frac{e}{k^2}\right)^{k^2}(r/\beta)^{2k^2}\exp(-r^2/\beta^2)$$

(3.29)

for $k = 0, 1, \ldots n_b$
where \( n_b \) denotes the number of basis functions included and \( \beta \) is a parameter that is of the order of annuli thickness. The individual bases of the BASEX set (Fig. 3.8) span the entire field variable domain uniformly with the maxima for each basis function at \( r = k\beta \), thereby allowing one to investigate how the location of an individual base inside the domain affects the reconstruction accuracy of an algorithm. The BASEX set also has an analytical expression for the projection of each basis function given by [79]

\[
\chi_k(x) = 2\beta \rho_k(x) \left[ 1 + \sum_{l=1}^{k^2} (x/\beta)^{-2l} \times \ldots \right.
\]

\[
\ldots \prod_{m=1}^{l} (k^2 + 1 - m)(m - 1/2)/m
\]

(3.30)

where \( \chi_k(x) \) denotes the projection of the \( k \)-th basis function. This provides an easy way to compare regularization and perturbation errors. Lastly, the BASEX set can also be easily used to recreate some of the more relevant property distributions (e.g. “Gaussian” or “top-hat” like distributions as shown in the insets of Fig. 3.13a and 3.13b ) generally encountered in the processes governed by diffusion e.g. in combustion.

### 3.2.2 Noise

The BASEX basis functions were combined together using coefficients generated with MATLAB’s sobolset generator thereby creating 10,000 pseudo-random test field distributions. The projections of
these pseudo random distributions were then corrupted by randomly distributed error i.e.

\[ p_{i,n} = p_{i,\text{exact}} + \text{er}(\mu, \alpha) \quad (3.31) \]

where \( \text{er}(\mu, \alpha) \) follows a normal distribution with mean \( \mu = 0 \) and standard deviation, \( \alpha \). The values of \( \alpha \) were chosen to range between 0.02 to 0.1 producing a maximum SNR (signal-to-noise ratio) in projections of the order of 35 to 20 db. The is reasonable since these are the typical level of noise often encountered in experimental setups such a TDLAS in combustion [refs]. These noisy projections were then used to test the precision and accuracy of reconstructions of various algorithms.

### 3.3 Results of Numerical Experiments

#### 3.3.1 Effect of DOE and Tikhonov Regularization

Abel inverted solutions suffer from two different kind of errors - regularization errors and perturbation errors. As mentioned before, regularization errors are only functions of the field to be reconstructed and the grid spacing. Therefore they affect only the accuracy (\( \varepsilon \)) of the solution. On the other hand, perturbation errors arise from the inherent ill-posedness of the problem when noise in projections are amplified in solution. Hence for a given grid spacing, the latter kind of errors affect the precision (\( \sigma \)) of the solution. Since the extent of ill-posedness depends solely on the projections or deconvolution matrix (\( A \) or \( D \)), which in turn depends on the ray / annuli spacings, an optimization procedure following the design-of-experiments theory [72] was performed to find the arrangement that is least susceptible to measurement noise for the 3 algorithms. Figure 3.9b shows the results from optimized ray distributions using the onion-peeling algorithm. The figure clearly shows that from the point of view of minimizing perturbation error, equi-spaced ray spacing is clearly not the optimal arrangement. Instead, a distribution with relatively larger ray spacing near the center with progressively smaller spacing near the edge of the domain is favored.

The optimization results can be explained by examining how the noise in the projections affect the noise in the solution. Figure 3.10 shows the variation of \( \delta f \) for unit rms fluctuation in measurements without any assumption of the field distribution, or spatial frequency of noise. As can be seen, for equi-spaced rays the fluctuations in the solution are small near the edge but gets increasingly worse
Figure 3.9: Variation of optimally spaced annuli edge location for OP, ATP and TPP as one approaches the center (bold lines in Fig. 3.10). It can also be seen, that as one approaches the center, the rate of increase of noise amplification also increases. From the above equation, it is obvious that the noise in solution is inversely proportional to $\Delta r$. Therefore it is no surprise that the optimization algorithm tries to keep annuli with large thicknesses towards the center. Figure 3.9a shows that characteristic “bow” shaped optimized distribution is common for all algorithms that have similar noise amplification characteristics. The TPP algorithm has a slightly different shape since the derivative at the center is calculated using a 3-point forward algorithm that uses the first two ray spacings near the center instead of just one.

It must be noted that design-of-experiments tries to maximize the information content of $\mathbf{A}$ (or $\mathbf{D}$) by changing the grid spacing. However, any change in $\mathbf{D}$ will have a corresponding change in $\delta \mathbf{D}$ thereby affecting the regularization error as well. Hence it is expected that depending on the function to reconstruct, regularization error may improve, remain unchanged or deteriorate with such an optimized arrangement. Since optimization produces lower resolution near the center but higher resolution near the edge of the domain it is expected that such a distribution is well suited for fields where most of the gradients are near the edge of the domain. Figure 3.12 which is a representative plot showing how the accuracy varies for reconstruction of different basis functions using the onion-peeling algorithm illustrates this point. For functions with all the gradients concentrated near the center ($f_1$ to $f_4$), the
equi-spaced rays perform much better whereas as the basis functions start moving outward ($f_5$ to $f_7$), the optimized ray arrangement produces better accuracy. Following this, two specific distributions that are often encountered in the combustion community were reconstructed. These were i) the “Gaussian” like distribution which is mathematically represented as

$$f_1 = (1 - r^2)^2$$  \(3.32\)

and ii) the “top-hat” like distribution, represented by

$$f_2 = \text{erf}(10 - 10r)$$  \(3.33\)

The former is a distribution that has most of the higher order gradients distributed near the center while the second is a distribution with almost all the gradients concentrated near the edge of the domain. Figure 3.13a and 3.13b show the precision and accuracy of reconstructions from equi-spaced and optimized distributions respectively. Each of these distributions have 10 rays and was contain 35 db of noise. The insets show the actual reconstructions and their uncertainties from the onion-peeling algorithm. It can be seen that for all the algorithms, the optimized arrangement performs better for the “top-hat” distribution than that for the “Gaussian” like distribution.
Figure 3.11 shows the effect of optimization on the singular value spectrum of the projection matrix. It can be seen, that the singular values of the equi-spaced arrangement (bold lines), for all 3 algorithms, have in general, a faster decay towards zero than the singular values for optimized rays (broken lines). Its effect of this change in singular value spectrum is immediately clear once we look at it in the context of Eq. 3.18. The second term in the equation denotes the amplification of measurement noise in solution scaled by the inverse of the singular values. A higher singular value therefore implies a smaller noise amplification in the solution.

In order to investigate the how reconstructions from the optimized arrangement compares with the equi-spaced beam arrangement a wide range of test distributions were reconstructed. 10,000 pseudo random distributions were generated combining coefficients from MATLAB’s “sobolset” generator with the BASEX basis functions. A typical cloud-plot of the reconstruction results using the TPP algorithm is shown in Fig. 3.14. The summary of the cloud plot results are shown in Fig. 3.15a which has been constructed using the centroids of the reconstruction clouds with two different levels of noise - 35 and 20 db. It can be seen that, irrespective of the algorithm, the noise level does not affect accuracy of reconstruction, but only the precision. While optimization positively suppresses noise from projections, resolution near the edge of the domain is increased at the cost of the resolution near the center. Hence depending on the distribution of gradients inside the function, the overall reconstruction may increase...
Figure 3.12: Regularization error (accuracy) for reconstruction of 7 basis functions with equispaced and optimized arrangements (onion-peeling algorithm)

decrease or remain unchanged. Comparing between the algorithms, one can see that although ATP is the best for noise suppression, TPP or onion-peeling tend to be more accurate.

Finally, reconstructions of the pseudo random functions from optimized rays, equi-spaced rays and Tikhonov regularized equi-spaced rays have been put in the same precision-accuracy plot (Fig. 3.15b) for comparison. As for Tikhonov regularized reconstructions, results from 3 levels of regularization i.e. with three different values of the regularization parameter $\lambda = 0.1, 0.063$ and 0.3 have been shown shown. For $\lambda = 0.1$ (green markers), the Tikhonov regularized reconstructions outperform the optimized reconstructions in both precision and accuracy for the OP and TPP algorithm. However, for ATP, the optimized rays still perform slightly better in terms of precision but fare worse in accuracy. However, with a slightly decreased level of regularization, $\lambda = 0.063$ (black markers), all the algorithms fare worse in precision as compared to the optimized system, though they still perform better in terms of accuracy. An effort to suppress the noise by increasing the regularization to $\lambda = 0.3$ produces reconstructions with higher precision (as shown by the magenta markers) but lower accuracy as compared to both the equi-spaced or optimized system for all algorithms. The result shows that Tikhonov regularized reconstructions have precision and accuracy that are strong functions of the regularization parameters as well as algorithms. This effect of regularization parameter on accuracy and precision is obvious - a higher
3.3.2 Effect of Averaging

To investigate the effects of sample averaging on reconstruction accuracy and precision, we consider a model distribution (also created using the BASEX functions) and its reconstruction using only the OP algorithm. This particular distribution has gradients that change magnitude and sign all throughout the domain and therefore serves well to highlight the different pros and cons of the strategies being discussed here – beam optimization, Tikhonov regularization and sample averaging. The measurement data set (i.e. set of projection values) is simulated and shown in Fig. 3.16a. Instead of a single mean value, the array of projection data is a set of 100 samples at each radial location. Each of these are randomly selected from a population of measurements corrupted by a 30db maximum noise level. Reconstructions using OP, but with no other error mitigation technique, is shown in Fig. 3.16b. A boxplot is used here to show the statistics of the reconstruction. The midpoint of the box is the median, the edges of the box are the 25th and 75th percentile respectively, while the whiskers extend up to the farthest data point that is not an outlier. Outliers are plotted individually. The open circular markers denote the mean of the reconstructed function values.

Figure 3.17 shows the same reconstruction using optimized ray spacings but no averaging. It is immediately obvious that the optimization serves well to reduce the spread ($\sigma$) of the reconstructed distribution reconstructions ($f_1$) and (b) Top hat distribution reconstruction ($f_2$)

regularization leads to a smoother or less noisy solution while simultaneously deviating away from more accurate solutions.
Figure 3.14: A representative cloud-plot from individual reconstructions of 10000 pseudo-random functions comparing the precision vs accuracy for optimized ray distribution (red) and equispaced ones (blue) function. On the downside, the accuracy of reconstruction is compromised near the center of the domain due to reduced resolution.

In the next figure, Fig. 3.18, the reconstructions are based on means of 100 sample sets, each sample set consisting of 10 samples. The benefits of averaging are reflected in the reduced spread of the reconstructed data around the mean. Figure 3.19, shows the same reconstruction when using a moderate value of the regularization parameter $\lambda = 0.3$. The figure shows that this method further reduces the $\sigma$ of the reconstructed distribution while hardly affecting the accuracy.

3.3.3 Concluding Remarks

The two different sources of errors in Abel inverted solutions - regularization error and perturbation error, were investigated in details in this chapter in the context of 3 different algorithms - OP, ATP and TPP. It was shown that irrespective of the algorithm, the regularization error is only a function of the local grid spacing and higher order derivatives of the field distribution or projection values and hence difficult to be completely ascertained a-priori. However, an approximate determination of regularization error can be carried out a-priori using the regularization coefficient and assuming a constant order of magnitude of the derivatives. Additionally, the regularization error (or accuracy of the solution) is not affected by the noise in projections. In contrast, the perturbation error (precision of the solution) is only a function
Figure 3.15: Centroids of cloud-plots for the 3 algorithms using equispaced, Tikhonov regularization and optimized rays to illustrate effect of increasing noise in projection and regularization parameter on accuracy and precision.

Figure 3.16: A simulated measurement data set for a projection containing random errors and its reconstruction by OP.
of the ray arrangement (the grid spacing) and error in measurements. Therefore, the perturbation error can be ascertained independently without any reference to the field variable or projection values. With regards to performance of algorithms, it can be concluded that although the ATP is less susceptible to noise in projections, the TPPa and TPPb tend to be more accurate for the given number of rays. Plotting the overall regularization error coefficient versus the overall noise coefficient revealed that for a given level of accuracy, the ATP algorithm is least prone to noise amplification.

Using flexible ray spacing, results from the design-of-experiments showed that equi-spaced rays are not optimal from the point of view of perturbation error minimization. In fact, the noise coefficient [38] which is extremely high near the center forces the optimization to favor arrangements that are well spaced near the center and closely spaced at the edge of the domain. The change in domain resolution from the center to the edge hence changes the regularization errors as well. Results from the reconstruction of the individual bases showed that the optimized arrangement is well suited for functions which have most of their gradients near the edge rather than distributions with significant higher order derivatives within the body or center of the domain.

Reconstructions from Tikhonov regularized inversion showed that results vary significantly based on the value of the regularization parameter. A value of $\lambda = 0.1$ with OP and TPP produced reconstructions that have better merit for accuracy and precision than optimized rays but was still worse in precision.
Figure 3.18: Reconstruction using equi-spaced rays and averaging.

Figure 3.19: Reconstruction using both Tikhonov and averaging (OP)
for the ATP. On the other hand, with $\lambda = 0.063$ all three algorithms produced better accuracy but worse precision than the optimized rays.

Finally, it was shown how simple averaging helps in reducing the $\sigma$ values of the reconstructed variables and can be an useful tool when used along with Tikhonov regularization. Previously, researchers in the field stayed away from closely spaced rays since even small residual errors after averaging would create a large variation in the solution. Averaging with sufficient number of sample sets, theoretically, would help in making the measured value of the projection $p_{\text{meas}}$ approach the ideal value of the projection. In such a situation one can use a high number of rays and Tikhonov regularization with a moderate value of $\lambda$ that helps promotes smoother solutions but without much detrimental effects on accuracy.

### 3.4 2D Tomography: Classical and Limited View Inversion

In 2D tomography where the simplifying assumption of axial symmetry is lacking, solution to the problem becomes increasingly complicated since number of unknowns increase geometrically. To illustrate the point, we take the case of the 1D tomography explained previously. Discretizing the domain into $N$ annular elements is sufficient to solve the problem using $N$ ray, but in the case of 2D tomography the domain (assumed as a square one for simplicity) needs to be discretized into $N$ parts in each direction ($x$ & $y$) thereby giving rise to $N^2$ pixels or unknowns. Also, to better sample the directional nature of the distribution, projections (or path integrals along the rays) need to be recorded from different angles. This is routinely done in medical imaging where CT-scan machines record projections from all around a patient’s body which are then transformed to the spatial frequency domain using the Fourier Slice theorem [80]. However, drastically reduced number of angular views leaves gaps in the recreated frequency description of the distribution and hence this technique fails when number of views are small. In such cases limited view tomography algorithms perform particularly well [43, 80]. These methods are briefly explained below.

#### 3.4.1 Filtered Back Projection (Classical)

This method is the classical method that produces tomographic reconstructions based on exact analytical expressions [41, 80, 81, 36]. The central idea behind FBP is that given the two-dimensional Fourier transform of an object, a reconstruction of the object can be performed by carrying out an inverse of the 2D-Fourier transform. The method by which 2D-Fourier transforms of an object are calculated
from its projections is known as the Fourier-Slice theorem (Fig. 3.20 [82]). The 1D-Fourier transform of a set of parallel Radon transforms [83] (or a set of parallel rays in a projection), inclined at an angle $\theta$ with a reference axis in the space domain, produces a spatial frequency distribution along a slice in the object’s 2D frequency distribution. In the 2D-frequency domain of the object this slice subtends the same inclination with the reference axis as in the space domain. Mathematically, the space domain description of the object, $f(x, y)$, and its frequency domain description $F(u, v)$, are related to each other as

$$f(x, y) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} F(u, v)e^{i2\pi(ux+vy)}dudv$$  

(3.34)

Interchanging between rectangular $(u,v)$ and polar $(w, \theta)$ co-ordinate system in the frequency domain, substituting $u = w \cos \theta$, $v = w \sin \theta$ and $dudv = wdw\theta$, and using the property $F(w, \theta + \pi) = F(-w, \theta)$, Eq. 3.34 is rewritten as

$$f(x, y) = \int_{0}^{\pi} \left[ \int_{-\infty}^{\infty} F(w, \theta)|w|e^{i2\pi wt}dw \right] d\theta$$  

(3.35)

where, $t = x \cos \theta + y \sin \theta$. Here, $F(w, \theta)$ is the Fourier transform of the object taken at angle $\theta$ which, – applying the Fourier Slice theorem, – is equivalent to $S_{\theta}(w) = \int_{-\infty}^{\infty} P_{\theta}(t)e^{i2\pi wt}dt$, where $P_{\theta}(t)$ denotes the projection inclined at $\theta$ with $x$-axis and $t$ distance from the origin. The quantity inside the square brackets in Eq. 3.35 is referred to as the “filtered projection”, whereas the term $|w|$ is the frequency response of the “filter”.

### 3.4.2 Limited View 2D Tomography

Unfortunately, optical access in experiments is often limited, which, together with cost of equipment, poses substantial problems in creating a tomographic measurement system that produces highly resolved directional information. Here 2D tomography with limited number of views is therefore approached as the solution of an inverse problem, where projection values are LOS integrated measurements of local absorbance characteristics. Two separate algorithms for the conversion of LOS integrated measurements (Eq. 2.36) to local values – MART and Tikhonov regularized inversion – use identical projection matrices, which convert local data to corresponding projection data obtained by LOS measurements. For the projection matrix, the domain of interest is discretized into a set of pixels (refer Fig. 3.21a). A ray, assumed as an infinitesimally thin beam, passes through the domain of interest, where its attenuated
Figure 3.20: A schematic explaining the use of the Fourier Slice Theorem in filtered backprojection. The projected value is detected by a transducer. The algorithms use these signals to reconstruct a discretized image. The intensity of the ray (or the projection value) is approximated as a weighted sum, i.e.

\[ \sum_{j=1}^{N} w_{ij} F_j = P_i, \quad i = 1, 2, 3 \ldots M \quad (3.36) \]

where \( N \) is the total number of pixels and \( M \) is the total number of rays. If each view contains \( m \) parallel rays and there are \( n \) views then \( M = mn \) is the total number of rays in the system. \( F_j \) is the local 2D value over the \( j \)-th pixel and \( w_{ij} \) is the normalized weight coefficient or fractional contribution of the \( j \)-th pixel to the \( i \)-th ray. The values of \( w_{ij} \) were approximated as the ratio of the length of the \( i \)-th ray intercepted by the \( j \)-th pixel to the total length of the \( i \)-th ray in the domain of interest. In matrix notation, the system of equation 3.36 becomes

\[ WF = P \quad (3.37) \]

As both \( W \) and \( P \) are known while \( F \) is unknown, Eq. 3.37 is an inverse problem. Since \( M < N \) and projections are contaminated by measurement noise, the inverse problem is ill posed, thus requiring advanced solution methods.
• MART

The multiplicative variant of the algebraic reconstruction technique (ART) [42, 43] used in the present study is adapted from Mishra, et al. [84]. In order to reconstruct individual 2f-peak height distributions via MART, the set of equations described in (3.36) is solved iteratively using an algorithm defined by

\[ F_{q+1}^{j} = C_{j}^{q} F_{j}^{q}, \quad j = 1, 2, 3...N \]  

(3.38)

where superscripts indicate iteration numbers. Elements of \( C_{j}^{q} \) - the multiplicative correction vector at the q-th iteration are computed as

\[ C_{j}^{q} = \prod_{i} \left( \frac{P_{i}}{< w_{i}. F_{q}^{q} >} \right)^{\mu w_{ij}} \]  

(3.39)

where \( \mu \) is a relaxation parameter and \( M_{i} \) is the number of rays that intersect the \( C_{j} \)-th pixel. In Equation 3.39, the inner product \( < w_{i}. F_{q}^{q} > \) denotes the approximation of the projection value based on the existing pixel values. Hence, in MART, the magnitude of the update due to the i-th ray is the ratio of the measured projection value \( P_{i} \) to the approximated projection value raised to the proper exponent. The exponent ensures that only those pixels that are intercepted by the i-th ray are updated. An iteration is completed when all the rays have been used up to find the total correction vector.

Any algebraic reconstruction technique suffers from local high frequency noise [80, 41]. This noise, also called 'salt-and-pepper' noise, is vestigial of the iterative correction technique that the MART method utilizes [80, 40]. To eliminate noise inherent to solutions obtained by MART, a two-dimensional median filter with a window of size 3 x 3 pixels was incorporated into the algorithm. Digital median filters are commonly used in image processing to remove 'salt-and-pepper' noise [85]. Digital median filters operate on each pixel individually by replacing it with the median value of a set that contains values from its neighbors. The size of the set is determined by the window size of the filter. Such a filtering technique though very useful for removing 'salt-and-pepper' type of noise is also expected to adversely affect the edges of a distribution with sharp gradients like the Tophat phantom. In addition to MART, median filtering was also used to reduce noise after point-wise reconstruction of temperature by two-line thermography.
Figure 3.21: Domain discretization and choosing the regularization parameter for Tikhonov regularization

- **Tikhonov Regularized Inversion**

The second algorithm used in the study is a Tikhonov regularized inversion for rank deficient problems. A comprehensive overview of the different methods of tackling such problems can be found in Hansen’s work [73]. The linear system $WF = P$ (the matrix form of Eq. 3.36), consists of $N$ unknown pixel values and $M$ rays. The rays are integral equations that relate the pixel values to the projections. Due to lack of optical access, engineering difficulty or just sheer high-cost, it is difficult to have a high number of rays inside any domain. Therefore, in general, $M < N$. This means that it is a rank-deficient problem without a unique solution. In the absence of a unique solution, an approach that is often used is to find an approximation that best explains the set of measured projections under the given constraints while simultaneously ensuring that the solution has a smooth distribution. In other words, a solution $F^{LS}$ is sought such that $\arg(||P - WF||)$ is minimized. The methodology of the solution followed here matches closely that of Daun and Twynstra, et al[78, 46]: due to the rank-deficiency, the solution set to this problem will not just contain the least square solution but also a non-trivial solution, $F^n$, belonging to the null-space of the problem [78, 73] i.e. $WF^n = 0$. To span this null space, the system is augmented by additional information that guarantees smoothness and non-negativity. This is done by applying Tikhonov regularization to the system of equations by adding another set of $N$ equations, $\lambda LF = 0$,
where \( \mathbf{L} \) approximates the discrete version of the ‘\( \nabla \)’ operator. Therefore,

\[
L_{ij} = \begin{cases} 
1 & \text{if } i = j \\
-1/k & \text{if } \text{neighbors}(j) \\
0 & \text{otherwise}
\end{cases}
\]  \hspace{1cm} (3.40)

where \( k \) is the number of neighbors of a pixel. The regularization parameter \( \lambda \), along with the additional set of equations shown in Eq. 3.40, rejects noise while preserving the structure of the unknown distribution. Thus, the problem is reduced to finding a solution as

\[
\mathbf{F}_\lambda = \arg \min \left\{ \| \mathbf{W}_\lambda \mathbf{L} \mathbf{F} - \begin{bmatrix} \mathbf{P} \\ 0 \end{bmatrix} \|_2^2 \right\} \text{ such that } \mathbf{F} \geq \mathbf{0} \hspace{1cm} (3.41)
\]

The success of this method hinges on the magnitude of the regularization parameter \( \lambda \). It is obvious that a low value of \( \lambda \) results in a highly “corrugated” reconstruction due to the low amount of smoothing via the matrix \( \mathbf{L} \); conversely, a large \( \lambda \) value results in a distribution that is too smooth and thus does not preserve the features of the actual distribution. One method of finding a proper value for the regularization parameter is using the “L-curve” criterion (LCC) [73]. However, in this work, we use the method explained by Daun, et al. [45]. In short, the singular values of the augmented matrix \( [\mathbf{W}^T \lambda \mathbf{L}^T]^T \) for different values of the regularization parameter are plotted. Rank-deficient problems, however, have a singular value spectrum that spans several orders of magnitude with a distinct gap in the spectrum (Fig. 3.21b), which results in insufficient smoothing. Daun, et al. [45] suggested a value of \( \lambda \) that pads the small singular values but does not overwhelm the large non-trivial singular values.

The solution \( \mathbf{F}_\lambda \) in Eq. 3.41 can alternatively be written as

\[
\mathbf{F}_\lambda = \mathbf{W}^\# \mathbf{P} \hspace{1cm} (3.42)
\]

where \( \mathbf{W}^\# \) is the Tikhonov regularized inverse given by [73]

\[
\mathbf{W}^\# = (\mathbf{W}^T \mathbf{W} + \lambda^2 \mathbf{L}^T \mathbf{L})^{-1} \mathbf{W}^T \hspace{1cm} (3.43)
\]
If $\delta P$ is the error in measurement of projections then following the procedure outlined for 1D tomography, it can be written that

$$P_{\text{meas}} = P_{\text{ex}} + \delta P = WF_{\text{ex}} + \delta P$$

(3.44)

Therefore,

$$F_{\lambda} = W^#WF_{\text{ex}} + W^#\delta P$$

(3.45)

which gives

$$\delta F = F_{\lambda} - F_{\text{ex}} = (W^#W - I)F_{\text{ex}} + W^#\delta P$$

(3.46)

Here the term $(W^#W - I)F_{\text{ex}}$ is the regularization error while the term $W^#\delta P$ is the perturbation error i.e. error due magnification of measurement noise. One of the chief differences of 2D tomography with 1D tomography is that the non-trivial singular values of $W$ are quite large and the solution $F_{\lambda}$ is almost invariant w.r.t. perturbation error over a large range of $\lambda$ [78]. Therefore, in 2D tomography, the perturbation error is negligible compared to the regularization error.

### 3.4.3 Concluding Remarks

Compared to 1D tomography, which is an ill-conditioned problem, the 2D tomography problem is different in the sense that it is a rank-deficient problem. In case of 1D tomography Tikhonov regularization is used to stabilize the solution in the presence of noise whereas for the 2D case the same is used to augment “missing information” with information of “smoothness” and “non-negativity” of the field. In 1D tomography, errors stemming from both regularization (non-uniform ray spacing or sparse spaced grid) and perturbation (noise in projections and/or fine grid resolution) may create amplified noise in solution while in 2D tomography only regularization errors are sources of major artifacts in the solution. Apart from individual errors in spectroscopic data reconstruction, it is as yet unknown whether the use of 2-line thermometry can create major artifacts in either case (1D or 2D). Hence the next chapter specifically analyzes the sources of “tomographic-TDLAS artifacts” using simulated data.
Chapter 4
Artifacts in Tomographic TDLAS

One of the major drawbacks in any inverse technique like tomography is the difficulty to differentiate between actual features of the distribution versus mathematical reconstruction artifacts that may arise due to the solution of a ill-conditioned (1D tomography) or a rank-deficient problem (limited view 2D tomography). Also, it needs to be ascertained if the use of two-line ratio TDLAS in conjunction with tomography creates additional artifacts in final temperature and concentration. In order to investigate this, analysis was carried out using simulated data for both 1D and 2D tomographic TDLAS. Sources for measurement noise are included through models for laser and photodetector noise [13]. Time series data for laser intensities along with photodetector shot and thermal noise were created following the procedure proposed by Lins et al. [13]. The modeled intensities were then used to calculate projection values which were subsequently used for tomographic reconstruction. It is worthwhile to mention that the noise models included in this work are not comprehensive enough to include all kinds of errors. However, it must be noted that the major source of noise in most TDLAS setup is the low-frequency flicker noise from the diode which has been included using models based on actual equipment noise data published by Lins et al. Noise due to beam-walking, window fouling etc. can be compensated in case of WMS-2f by use of 1f normalization of 2f peak heights. Other noise sources like back-reflections from optical components or flow variations, while important, are also difficult to be accounted for using simulation models. The main reason to model noise in this study is to investigate its contribution to the creation of TDLAS artifacts.

4.1 Simulated Tomographic TDLAS Artifacts in 1D

4.1.1 Data Set

This work reconstructs the temperature and water vapor concentration profiles of a stationary test flame with properties that correspond to published time-averaged values of a Sandia D flame [86]. The values used are values taken at a horizontal section which is 432 mm above the burner nozzle. Temperature and concentration profiles are illustrated in Figure 4.1. The projection data is obtained from simulated measurement values of line-of-sight spectral absorbance (DA), or the second harmonic peak...
Figure 4.1: Temperature and concentration distributions of the simulated flame. Distributions comprise of time averaged values taken from a section of the Sandia D flame (at a height-above-burner = 432 mm).

heights of the transmitted beam intensity (WMS-2f). These simulated projection data are then processed by the Abel inversion method to reconstruct the local spectral absorbance and 2f peak height values throughout the domain.

The distributions of spatially resolved spectroscopic data based on local temperature and concentration values are shown in Figure 4.2. For both direct absorption (DA) and wavelength modulation (WMS-2f), local temperatures (varying from 300K to 1940 K) and concentrations (varying from 0 to 8 mol % of water vapor) create characteristic distributions for each of the two transition lines: the distribution of spectroscopic data for line 1 (6931.59cm$^{-1}$) resembles a Gaussian curve, whereas the distribution for line 2 (6935.98cm$^{-1}$) resembles a ‘top-hat’ profile (Fig. 4.2). The different characteristics stem from the fact that spectral absorbance values are non-linear functions of temperature, determined by line-strengths $S_i(T)$ for different transitions. While Figure 4.2 illustrates exact distributions of spectroscopic data, reconstructions using Abel inversion and FBP infer those distributions from projected data. Thus, these distributions serve as benchmark/target data that are used to assess the quality of reconstruction of spectral absorbance for DA (Fig. 4.2a) and 2f peak heights for WMS-2f (Fig. 4.2b).
Figure 4.2: Spatially resolved spectroscopic benchmark data for two H$_2$O absorption lines. Tomographic reconstruction involves recreation of integrated absorbance for direct absorption (a), and 2f-peak heights for WMS-2f (b).

4.1.2 Reconstructions

The Abel inversion is based on axially symmetric property fields. Simulated projection values are obtained for parallel rays passing through the test flame from a single direction, and radially resolved spectroscopic data are reconstructed from line-of-sight projections using Dasch’s 3-point algorithm[38]. In the case of direct absorption spectroscopy (DA), the Abel inversion yields the distribution of integrated absorbance values for the two H$_2$O transitions. In the case of wavelength modulation spectroscopy (WMS-2f), the reconstruction involves 2f peak heights. In either case, two sets of spectroscopic data are used to obtain radially resolved distributions of temperature and concentrations.

Direct Absorption Spectroscopy (DA). Reconstructed values of normalized integrated absorbances for the two transition lines are shown in Figure 4.3a. The top panel illustrates that the radial distribution of integrated absorbance values is reconstructed correctly. The bottom panel shows the deviation of reconstructed values from target data as well as estimated uncertainty of the reconstructions introduced by the laser and photodetector noise. For cases with added noise, data from 100 scans were averaged and spline fitted before deconvolution.\footnote{Marginally better results are expected for Voigt profile fitting, which, however, requires reversing the order of fitting and tomographic reconstruction, introducing significant computational overhead.} While deviations in reconstructions remain within 2% of the target values, the uncertainty level due to noise is about 50% of peak values. Thus, reconstructions of DA data
Figure 4.3: Reconstruction of spectroscopic data using Abel inversion: reconstructed integrated absorption signals for DA shown in (a) and reconstructed 2f peak heights for WMS shown in (b).

Figure 4.4: Reconstruction of temperature and concentration distributions using Abel inversion.
containing noise are rendered meaningless, which is mainly due to the fact that the absorbances for the chosen line pair are relatively weak.

**Wavelength Modulation Spectroscopy (WMS-2f).** In comparison to DA, Figure 4.3b clearly illustrates the powerful noise reduction capability of the WMS-2f technique. While the quality of reconstruction of data without noise is comparable to DA, the uncertainties due to noise are drastically reduced, and remain below the errors incurred due to the Abel inversion. The deviations of the reconstructions are most pronounced at \( r \approx 40 \), where temperatures and concentrations are low. Further outside, deviations are reduced, as the inversion algorithm performs well at approximating values that go to zero with a zero gradient[53].

**Temperature and Concentration Reconstructions.** Figure 4.4 shows simulated measurements of radial temperature and concentration distributions of the test flame based on reconstructed values of spectroscopic data (Fig. 4.3). Results clearly illustrate that the Abel inversion yields sufficiently accurate spectroscopic data to accurately reconstruct temperature and concentrations using both spectroscopic techniques. Without laser noise, deviations for temperatures and concentrations remain below 25 degrees and 0.25 mol %, respectively. A comparison between the temperature and concentration results obtained from DA and WMS-2f reconstructions shows that the reconstructions follow similar trends, although WMS-2f results fare slightly better than DA results, especially for concentrations. This is due to the fact that reconstruction errors in temperature are propagated to concentration results due to the temperature-dependent line-strength term \( S(T) \) in Eq. 2.28. This, coupled with reconstruction errors in integrated spectral absorbance \( A_i \), produces a larger error in the DA results.

The impact of noise on reconstructions involving WMS-2f is illustrated in the bottom panels of Figure 4.3. For WMS-2f, the uncertainty due to noise is low at the center and rises considerably as temperatures and concentrations decrease. This is mostly due to the fact that the absorption signal near the edges is too weak and thus easily corrupted. However, average values from WMS-2f remain almost indistinguishable from noiseless data. This result is in stark contrast to DA data, where the large amount of noise in the spectroscopic data – shown in Fig. 4.3a – renders results meaningless. As uncertainties in temperature and concentration reconstructions would be unacceptably high, corresponding uncertainty bars have been suppressed for DA data.
4.1.3 Observations: Reconstruction Artifacts and Noise

This study aimed to illuminate the impact of tomographic artifacts and laser noise on the quality of in-situ measurements of temperature and concentration distributions by tomographic TDLAS of a simulated test flame. In particular, the performance of two spectroscopic techniques – direct absorption spectroscopy (DA) and wavelength modulated spectroscopy (WMS-2f) – was tested for Abel inversion. The tomographic deconvolution of simulated projection data creates spatially resolved spectroscopic data, which is used to calculate local values of temperature and concentration. Results for simulations without modeled noise show that both spectroscopic techniques perform well for the reconstruction of temperature, although WMS-2f shows slightly better results than DA for concentrations. If laser noise is included, simulations show the expected outcome that WMS-2f outperforms DA due to its inherent noise reduction capabilities. Hence, in the context of 1D tomography, it is observed that artifacts from reconstruction error are not significant enough. Although laser noise itself can mar reconstructions results from DA if transitions are not strong enough, WMS-2f is ideal to suppress any detrimental effects of laser noise which can ruin tomo-synthesis results. However, it should be noted that the laser noise is not the only kind of noise present in an experimental setting and hence Tikhonov regularization, which has been shown produce superior results to existing Abel inversion algorithm for inversion of data corrupted with experimental noise [68], will be used for 1D tomography.

4.2 Simulated Tomographic TDLAS Artifacts in 2D: Filtered Back Projection

4.2.1 Data Set

Similar to the simulated work carried out in 1D tomography to gauge the effect of tomosynthesis artifacts on reconstructed temperature and concentration, the filtered back projection algorithm was used to reconstruct temperature and concentration values of the same phantom flame (Sandia D), except that the center of the flame was offset from the center of the domain by $\Delta x = \Delta y = 10$ units.

Filtered backprojection (FBP) can be applied for the reconstruction of non-symmetric property fields. Compared to the Abel inversion, FBP requires a significantly increased amount of data for high-quality reconstructions. While recent iterative tomographic methods like ART or MART are generally better suited for reconstructions based on a limited number of views, the reconstruction quality is affected by the choice of parameters of the algorithm [42, 43], and filters required for the removal of 1salt-
Figure 4.5: Evolution of the test distributions across number of views using WMS-2f and FBP. Top two rows (a and b) are normalized 2f distributions. The bottom two rows (c and d) are temperature and concentrations.

and-pepper’ noise[80]. In comparison, FBP produces well-defined and mathematically well-understood tomographic artifacts that are a function of property distribution, number of views and number of rays per view (rpv) only. A detailed review of artifacts occurring due to undersampling of angular views is given by Kak et al.[80]. In the following, all reconstructions were obtained for 15 views and 152 rays per view (ray spacing of 1.1 units) using a standard ‘Ram-Lak’ [80, 81] filter for the projections. While the impact of rpv on reconstruction quality was investigated, no significant variation of results was observed above 51 rpv (3.3 unit spacing).
4.2.2 Reconstructions

Figure 4.5 illustrates the impact of number of views on the reconstructions of spectroscopic data and corresponding simulated temperature and concentration distributions for the WMS-2f method. Results clearly illustrate substantial artifacts for limited numbers of views – 2, 3, and 5, – where the number of characteristic radial artifacts is twice the number of views. The two top rows of Figure 4.5 clearly show the diminishing effects of reconstruction artifacts in spectroscopic data with increasing number of views; at 15 views, the magnitudes of artifacts are significantly reduced, and results approach the benchmark. The two bottom rows of Figure 4.5, however, illustrate that artifacts present in temperature and concentrations are significantly larger than those observed in the reconstruction of spectroscopic data. Again, at 15 views, results are significantly improved, although some artifacts around the perimeter of the flame are clearly visible. The increased prominence of artifacts in the reconstruction of temperature and concentrations is due to the discrepancy of artifacts in the reconstructions of 2f peak heights (WMS-2f) for the two absorption lines. It is clearly visible that the extent of the peak for line 2 is larger than that for line 1, which produces noticeable differences of artifacts outside of the flame zone. Those differences propagate into the calculation of temperatures and concentrations, where the magnitudes are amplified significantly. While results are shown for WMS-2f only, simulations using DA spectroscopy yield similar results, although the magnitude of artifacts is somewhat increased.

Reconstruction of Spectroscopic Data. In order to further investigate the impact of tomographic artifacts on the reconstruction quality, radially averaged results are calculated over one quarter of the reconstructed features ($x \leq 10$ and $y \leq 10$), i.e. a radial distance $r = 0$ corresponds to the center of the features, whereas values for $r > 0$ are averaged over a quarter circle. Due to tomographic artifacts, values are non-uniform, and the standard deviation of local values is used as a metric of their magnitudes.

Figure 4.6 illustrates results for radially averaged integrated spectral absorbances (DA) and 2f peak heights (WMS-2f). Similar to results from Abel inversions, FBP yields accurate reconstructions of the spectroscopic data, where local deviations remain below 1% as illustrated in the bottom panels. The magnitude of deviations due to artifacts at a particular radius from the flame center is indicated by error bars. Comparing results from DA to results from WMS-2f, it can be seen that although deviations of mean values are comparable, the magnitude of artifacts at $r \approx 40$ is higher for DA, whereas it is higher for
WMS-2f outside of the flame. Overall, Figure 4.6 clearly depicts that even without the presence of measurement noise, the reconstruction of WMS-2f heights is less prone to artifact errors than reconstructed values of spectrally integrated absorbance for DA.

Reconstruction of Temperature and Concentration. Figure 4.7 illustrates temperature and concentration results for both DA and WMS-2f. Similar to the impact of laser noise in Abel inversion, in case of 2D tomography, results clearly indicate that artifact errors negatively impact the reconstruction quality as temperature and concentrations decrease. In the central region, WMS-2f performs slightly better than DA. Close to the edge of the flame, however, values reconstructed by DA become meaningless, whereas WMS-2f results deteriorate at $r \approx 40$, before they recover outside of the flame zone. The intermittent deterioration of WMS-2f is attributed to differences in the absorption characteristics between the two lines, where the 2-f peak heights approach zero at a different radial distances. Presence of small reconstruction artifacts in these regions therefore create significant errors in final values.
Figure 4.7: Reconstruction of temperature and concentration distributions by WMS-2f and FBP from 15 views and 152 rpv. Error bars indicate non-uniformity of results at a radial distance.

Increase of uncertainty at the perimeter of the flame \((r \approx 50)\) are observed in both Fig. 4.7 and Fig. 4.4 in the region where temperatures and concentrations are low. While the sources of uncertainty are different, – measurement noise and tomographic artifacts, respectively, – they are caused by the same mechanism. In both cases, small errors are introduced in reconstructed spectroscopic data (Figs. 4.3/4.6). These relatively small errors are, however, magnified by the spectroscopic technique, which deteriorates whenever temperatures and concentrations are calculated based on weak signals. It is noted that the two absorption lines approach zero at different rates as the radius increases (Fig. 4.2). As this discrepancy introduces different patterns in tomographic artifacts in spectroscopic data for the individual lines (Fig. 4.5), it is concluded that the choice of absorption lines affects the quality of tomographic reconstructions.

4.2.3 Observations: Effect of Artifacts

This work seeks to illuminate the impact of tomographic artifacts on the quality of in-situ measurements of temperature and concentration distributions by tomographic TDLAS of a simulated test flame.
In particular, the performance of two spectroscopic techniques – direct absorption spectroscopy (DA) and wavelength modulated spectroscopy (WMS-2f) – was tested using the classical tomographic reconstruction technique of filtered backprojection. The tomographic deconvolution of simulated projection data creates spatially resolved spectroscopic data, which is used to calculate temperature and concentration of the flame locally. Results for simulations without modeled noise show that both spectroscopic techniques perform well for the reconstruction of temperature, although WMS-2f shows slightly better results than DA for concentrations. If noise is included, simulations show the expected outcome that WMS-2f outperforms DA due to its inherent noise reduction capabilities.

For reconstructions of non-symmetric domains using filtered backprojection, spectroscopic data for individual transition lines was reconstructed with good accuracy as long as a sufficient number of views was used. Yet, temperature and concentration reconstructions showed significant artifacts near the edge of the flame. The artifacts are attributed to different absorption characteristics for the two absorption features. Large reconstruction errors are attributed to the way temperature and concentration are calculated from spectroscopic data, where relatively small errors are magnified whenever local absorbance values are small. Thus, it is concluded that accurate temperature and concentration reconstructions from tomographic TDLAS require a careful selection of absorption lines, as differences in line characteristics in weakly absorbing zones can magnify tomographic artifacts in final temperature and concentration reconstructions.

4.3 Simulated Tomographic TDLAS Artifacts in 2D: Limited View Tomography

4.3.1 Domain Geometry and Test Phantoms

This work uses a square geometry of $60 \times 60$ pixels for the reconstructions of simulated test phantoms. Synthetic projection values for test phantoms were sampled from the Radon space of corresponding high resolution ($800 \times 800$ pixel) phantom images. Two stationary phantom flames were considered: a 'Top-Hat' phantom and a 'Cos-Gauss' phantom. The top-hat phantom assumes a temperature and concentration distribution that resembled a simple off-center top-hat distribution (Fig.4.8a), i.e.

$$\text{Tophat}(x,y) = 0.5 + 0.5 \text{erf}[0.4 \{15 - R(x,y)\}]$$  \hspace{1cm} (4.1)
where R denotes the distance of a pixel from the center of the top-hat which have been assumed to be at +7.5 units in the x and y directions.

The second phantom was assumed to be a weighted superposition of two Gaussian peaks centered at 
\((x_1 = 0.2, y_1 = 0.1)\) and 
\((x_2 = 0.2, y_2 = -0.35)\) and a cosine peak (Fig. 4.8b) [43]. Expressing a cosine phantom as

\[
\cos(x, y) = \begin{cases} 
\frac{1}{4} \left[1 - \cos \left(2\pi \left(x + \frac{1}{2}\right)\right)\right] \times \ldots \\
\ldots \left[1 - \cos \left(2\pi \left(y + \frac{1}{2}\right)\right)\right] & |x, y| < \frac{1}{2} \\
0 & \text{otherwise}
\end{cases}
\] (4.2)

the CosGauss phantom can be written as

\[
\cosGauss(x, y) = \zeta [0.3\cos(x, y) + 0.8\exp\{-9(x - x_1)^2 - \\
\ldots [6(y - y_1)]^2 + \exp\{-8(x - x_2)^2 - [30(y - y_2)]^2\}] ]
\] (4.3)

where \(\zeta = 1.09\) is a normalizing factor. Therefore, the temperature and the concentration of the flame is given by

\[
T(x, y) = 300 + 1600 \text{ Function} \quad \& \quad X(x, y) = 0.18 \text{ Function}
\] (4.4)

where 'Function' can either be 'Tophat' or 'CosGauss'. The quality of reconstruction is assessed using
the RMS error norm

\[ \sigma = \frac{1}{N} \sqrt{\sum_{j=1}^{N} (F_{j}^{\text{recon}} - F_{j}^{\text{actual}})^2} \]  \hspace{1cm} (4.5)

In general, the reconstruction quality is affected by two types of errors - artifacts (or reconstruction error) and measurement noise error. To bring out the effect of these errors on the overall reconstruction quality by the two algorithms, error norms with and without measurement noise were plotted and analyzed for each case.

4.3.2 Phantom Reconstructions and Discussions

Due to different lower states energies, the two absorption lines form distinct 2f-peak distributions. Reconstructions of these 2f-peak heights for both transitions are carried out for 3, 5 and 10 views with each view containing either 40, 60, 90 or 120 parallel rays per view. Each view was assumed to subtend equal angle with its adjacent view and rays inside each view were also assumed to be equi-spaced. It is known that the orientation of the view arrangement with respect to the domain has a significant bearing on the quality of reconstruction [87, 88]. As the present study focuses on the comparative merits of the two reconstruction algorithms, the orientation for each of the view-arrangement was fixed to the orientation with optimal reconstruction, where in each case, identical orientations were used for both tomographic techniques.

This work uses views that subtend equal angles among themselves. However, the orientation of a view w.r.t the principal axes of the domain also plays a significant role in determining the quality of reconstruction since a change in orientation would mean a change in the values of the weighting matrix of the rays. Fig. 4.9 shows the variation of the error norm as the orientation angle for a view arrangement is changed. It can be seen that the configuration that produces the worst qualities coincide for both algorithms for all number of views used, irrespective of the distribution (Top-Hat or Cos-Gauss) under consideration. In other words, one would do well to stay away from such configurations. In contrast, the best configurations for reconstructions not only seem to be a function of the algorithm being used but also a function of the distribution being reconstructed. Fig. 4.10 shows the best and worst reconstructions of a Top-Hat phantom. The rest of the work presented here is based on orientation configurations that are neither the best nor the worst for each algorithm. In other words, they are results from representative configurations.
Figure 4.9: Error norm variation with orientation (60 rpv). Top row: 3 views, middle row: 5 views, bottom row: 10 views.
Figure 4.10: Best and worst reconstruction of the top-hat phantom with MART and Tikhonov due to variation in orientation of views
Fig. 4.11 summarizes the findings for the reconstruction error of the 2f-peak height distribution for both lines of the Tophat phantom. As results for the CosGauss phantom are comparable, they are not shown. It can be seen that for both algorithms, the quality increases drastically with increase in number of views from 3 to 5, which is expected. However, by increasing views from 5 to 10, MART does not gain any significant advantage whereas Tikhonov regularized inversion still seems to attain appreciable quality improvement. Also, for low number of rays per view ($\leq 60$), Tikhonov inversion performs significantly better than MART for all number of views. Beyond 60 rays per view, the inclusion of additional rays proves beneficial for MART, but does not further improve results from Tikhonov inversion. For Tikhonov inversion, adding more number of rays per view only helps when there are not enough number of rays per view to intersect every pixel - hence the increase in quality from 40 to 60 rpv; once there is sufficient information from a specific direction, additional rays do not further improve the results. Mathematically, a system with a high number of closely spaced parallel rays intersecting the same set of pixels will only increase the number of linearly dependent rows in the weight coefficient matrix $W$ and thus create more problems for the inversion. Comparing the quality of unperturbed reconstructions (dotted lines in Fig. 4.11a and b) to the quality of the solution obtained from data contaminated by noise (bold lines in the same figure), it is clear that Tikhonov inversion is only marginally affected by noise, whereas the same is not true in the case of MART. The superior noise-suppression ability of Tikhonov has been shown by
Figure 4.12: Evolution of line reconstruction (60 rpv, 30 db noise)
Figure 4.13: Temperature and concentration reconstructions for the two phantom flames with Tikhonov and MART (5 views, 60 rpv w/ noise)
Twynstra, et al. [46]. On the other hand, the median filtering incorporated into MART suffers from the drawback that the choice of the filter is completely empirical and is therefore dependent on the discretion of the user. Since Tikhonov inversion has a very well-defined method of choosing the regularization parameter this dilemma is avoided.

Fig. 4.12 shows the evolution of the reconstruction of 2f peak height of line 2 with number of views with each view containing 60 rays per view. For the Cos-Gauss phantom, it can be seen that Tikhonov regularization is more effective in capturing both the Gaussian peaks as compared to MART, for all views. Additionally, it is noted that the presence of artifacts is more prominent for reconstructions using MART than for Tikhonov reconstructions, particularly for low number of views. A similar observation can be made for the Tophat phantom, where the Tikhonov algorithm captures the thin annular edge of the Tophat better than MART.

The spectroscopic line (transition) (i.e. WMS-2f peak height) distribution reconstructions for 5 views and 60 rays per view and 30 dB noise were selected for determining temperature and concentration reconstruction using 2-line TDLAS theory (Fig. 4.13). Small artifacts in line reconstruction can lead to large errors in computation of 2f-peak ratio particularly in the low-signal areas [61]: ratio-thermometry uses the ratios of individual values from the two line reconstruction to determine temperatures for an individual pixel; based on temperature results, concentrations are determined in an iterative fashion. As a result, the temperature and concentration reconstructions generally contain more artifacts than individual line reconstruction, which are particularly strong where values are small and errors are amplified. To suppress artifacts, a $3 \times 3$ median filter was applied to the reconstructed temperatures and concentrations. It is important to note that median filtering only mitigates artifacts, but does not include any information that is inherent to the data. Due to the blurring effect of the median filter, the relative merits of MART and Tikhonov reconstructions are less clear than in the case of line reconstructions. The temperature and concentration reconstructions show that the Tikhonov algorithm performs better in reconstructing the Tophat phantom temperature distribution. Although the MART reconstruction of temperature and concentration of the core section of the flame is satisfactory, it is marred by the presence of severe artifacts in the temperature map particularly in the low signal areas. This was not observed in the Tikhonov inverted reconstructions. On the other hand, compared to the Tophat phantom, the Cos-Gauss phantom
flame is a significantly intricate distribution. Hence reconstructions of this phantom, seems to be only moderately well approximated by both the algorithms.

4.3.3 Concluding Remarks

Two limited-view tomography methods were tested for their relative performance in reconstructing the temperature and concentration distribution of two stationary phantom flames based on TDLAS. Line-of-sight (LOS) integrated synthetic WMS-2f measurements were de-convoluted using either a) Tikhonov regularized inversion or b) multiplicative algebraic reconstruction technique (MART), and local values were used for determining temperature and concentration of water vapor in the flame. It was found that for low number of views, the quality of reconstructions of absorption features was significantly better for Tikhonov reconstructions. It was also seen that measurement noise adversely affects MART, whereas Tikhonov was largely unaffected. Tikhonov regularization generally requires a smaller number of rays per view (rpv): e.g., reconstructions from noisy projection data from 5 views of comparable quality required 60 rpv for Tikhonov regularization and 90rpv for MART. While reconstruction quality of individual lines for MART improved significantly with increasing rays per view, no appreciable improvement was noticed for Tikhonov. Conversely, an increased number of directional information (views) appeared to benefit Tikhonov more than MART. In spite of these advantages in case of individual line reconstructions, Tikhonov regularized inversion did not significantly outperform MART in temperature and concentration reconstructions. This is due to fact that small errors in individual line reconstructions can lead to large errors while taking ratios of pixel values for the two lines. Since values of temperature and concentration computed through two-line ratio thermometry depend on these ratios, artifacts are inevitable in the final temperature and concentrations reconstructions for both these algorithms thereby creating inferior final results as compared to individual line reconstructions. The application of median filter, though useful for removing local 'salt-and-pepper’ noise, cannot rectify ratios affected by tomosynthetic errors. Therefore use of ratio-thermometry for tomographic TDLAS requires significant improvement in individual line reconstructions and/or improved techniques for the calculation of temperatures and concentrations from distributed WMS-2f data by alternate filtering and regularizing techniques.
Chapter 5
Experimental Setup and Results

This chapter contains information regarding choice of spectral transitions used for the work, methodology for their selection, method of characterization of the laser diodes, description of the experimental setup being used for 1D tomography, mathematical tools used to measure second harmonic peak heights of different transitions and methods to convert measured 1f-normalized 2f peak heights to temperature and concentration values. Finally, the 1D tomography results for individual line-reconstruction and determination of temperature and concentration are presented. The 1D results are then extrapolated to 2D and reconstructions are performed using 5 views, 60 rays per view and Tikhonov regularization. Results from 2D tomography are then benchmarked against results from 1D tomography.

5.1 Spectroscopic Transition and Laser Selection

In order to successfully use TDLAS for temperature and concentration measurement, proper selection of laser and absorption transition is crucial. Criteria for selection of absorption transitions for TDLAS measurements have been covered comprehensively elsewhere [3, 60, 24] and hence will be briefly re-explained here. Since it is required to determine the temperature and concentration of water vapor molecules in a flame, it is desired that we select a wavelength region, where absorption is only from water vapor and not from other gases in the flame like carbon-dioxide or carbon monoxide. The HI-TRAN 2008 and HITEMP 2010 databases are used for this. Figure 1.2 in chapter 1 shows the absorption band for three of the major species generally encountered in flue gases. This was simulated using spectroscopic parameters listed in the HITEMP 2010 database. It can be seen that around the 1.4μm (7143 cm\(^{-1}\)) region, water vapor (the species of interest for this work) has little interference from the other gases. Hence this zone was further explored for preferred transitions. Some of the important points while selecting spectroscopic transitions are as follows:

i) The difference in lower state energies between the two selected transitions needs to be high which ensures high temperature sensitivity. For DA spectroscopy where the temperature is directly ascertained from the ratio of the integrated absorbances of the two transitions, temperature is given by Eq. 2.27.
Differentiating this equation yields the temperature sensitivity for the line pair:

\[
\frac{dR}{R} = \frac{hc}{k} \frac{(E_1'' - E_2'')}{T}
\]  

(5.1)

Therefore, it can be seen a higher difference in line-pair lower state energies will mean a higher sensitivity for temperature.

In case of WMS-2f, temperature sensitivity of the two transitions can be calculated numerically from simulations of 2f peak heights variations with temperatures. In this case, a higher difference in lower state energies translates to a higher slope of the 2f-ratio vs temperature curve (refer Fig. 5.1) which is used as a “look-up-table” for determining temperature. This means a given percentage of error in finding the 2f-peak ratios leads to only a small error in finding temperature if the curve is steeper. As a rule of thumb a sensitivity value > 1 is preferred over the whole operating range. A energy separation of ~800 to 1000 cm\(^{-1}\) (note: it is customary in spectroscopy to express energy levels and their differences in terms of wavenumbers (frequency), \(\nu\) (cm\(^{-1}\)), using the relationship \(E = h\nu\) ) has been suggested in these literature for ensuring this. However, if operating temperatures are higher, then for obvious reasons a larger \(\Delta E\) is preferable. Selection of lasers are also contingent upon other practical factors such as availability of lasers and availability of fibers etc.
ii) The values of lower state energy levels for individual transitions are desired to be high as well. Since the species being probed here is water vapor (which is present in significant amounts in ambient atmosphere) and because of the fact that the laser arrangement used here contains significant path length outside the burner itself this criteria is important to avoid absorption from water vapor outside combustion zones. This ensures that most of the absorption takes place in the high temperature zones or the combustion zones and not outside.

iii) Minimum interference from neighboring transitions is desired. An isolated transition helps in data processing for obvious reasons since there will be no need for incorporating corrections.

iv) If possible, the transitions need to be close enough to be accessed by a single laser. Tunability of diode lasers are generally in the range of ~2 cm\(^{-1}\) for telecom grade diode lasers. This is a difficult criteria to satisfy since the choice of lasers are not only limited by the transitions available but also by the availability and affordability of diode lasers which can vary significantly even inside the telecom band, particularly if the transitions selected need to be custom manufactured.

Based on the above criteria, two transitions were selected viz. the transitions at 1442.67 cm\(^{-1}\) with a lower state energy of ~1800 cm\(^{-1}\) and the transition at 1391.69 cm\(^{-1}\) with a lower state energy of ~1000 cm\(^{-1}\). In order to keep the setup applicable for future use at higher wavelengths, free-space lasers were preferred over fiber coupled lasers. The other spectroscopic parameters for the chosen transitions have been taken from the HITRAN / HITEMP database[57, 20]. It should be noted here that for the lasers chosen here, the sensitivity value is < 1 for any temperature above ~1400 K (refer Fig. 5.1). Since the temperatures encountered even in a lean premixed flat-flame are well above 1400 K, this is obviously not ideal. Primarily, constraint with regards to laser availability dictated such a decision.

5.2 Lock-in-Amplifier (LIA)

Determination of temperature and concentration via the TDLAS theory requires the LOS 2f lineshapes to be de-convoluted to produce local values. To capture the second harmonic of the detector signal a lock-in-amplifier was created using LabVIEW. The raw detector signal is fed into the LIA which produces the LOS integrated 2f-peak heights at each radial location required for the tomographic de-convolution. A comprehensive explanation of how a LIA works can be found in the article by Scofield [89], therefore only a brief explanation is provided here. This is also schematically explained in the Fig. 5.2.
Let us assume a signal which can be analytically approximated as

\[ I = A_0 + A_1 \sin(\omega_1 t + \phi_1) + A_2 \sin(\omega_2 t + \phi_2) + \ldots + \text{noise} \]  

(5.2)

and we wish to find out the unknown amplitude \( A_1 \) oscillating with the known frequency \( \omega_1 \). To achieve this, the LIA multiplies the signal with a reference sinusoidal signal (R) of the same frequency, thus, \( R = \sin(\omega_1 t) \). Hence the output signal becomes

\[ S = RI = A_0 \sin(\omega_1 t) + A_1 \sin(\omega_1 t + \phi_1) \sin(\omega_1 t) + A_2 \sin(\omega_2 t + \phi_2) \sin(\omega_1 t) + \ldots + \text{noise} \times \sin(\omega_1 t) \]  

(5.3)

The second term in the expression can be simplified into \( \frac{A_1}{2} \left[ \cos \phi_1 - \cos (2\omega_1 t + \phi_1) \right] \) thus producing a DC component while all the other terms will have a harmonic attached to it. So, if the output of the LIA is low-pass filtered using an appropriate cut-off, then the resultant will be the x-component of the LIA given by

\[ S_{x_{\text{lia}}} = \frac{A_1}{2} \cos \phi_1 \]  

(5.4)

Similarly, multiplying the signal with a cosine reference will produce the y-component of the LIA given by

\[ S_{y_{\text{lia}}} = \frac{A_1}{2} \sin \phi_1 \]  

(5.5)

Here, the phase shift \( \phi_1 \) is also known as the detection phase shift of the lock-in-amplifier. This denotes
by how much the detector signal lags or leads the reference signal. One can adjust this to zero by adding
a phase shift in the reference signal \( R \) and it is useful to find out the individual \( x \) and \( y \) components (this
is also the reason for which lock-in-amplification is generally known as phase-sensitive detection \([89]\)).
One can avoid this cumbersome method of adjustment by finding the amplitude as

\[
A_1 = 2\sqrt{(Sx_{\text{lia}}^2 + Sy_{\text{lia}}^2)}
\]

(5.6)

Here, contribution from the noise is assumed to be canceled since the phase of noise is assumed to be
randomly distributed. Also, in the context of WMS-2f, with high modulation frequencies, noise levels
are significantly lower. Different harmonics of the detector signal is found by changing the modulation
frequency of the reference signal.

For WMS-2f, the detector signal which can be expressed by

\[
G_l(\nu) = GI_0(\nu)\tau(\nu + a\cos\omega t)
\]

(5.7)

will produce a LIA output whose \( X \) and \( Y \) components can be expressed as

\[
Sx_{\text{lia}} = \frac{G_l}{2}\left[H_2 + \frac{i_0}{2}(H_1 + H_3)\cos\psi_1\right]
\]

(5.8)

and

\[
Sy_{\text{lia}} = -\frac{G_l}{2}\left[\frac{i_0}{2}(H_1 - H_3)\sin\psi_1\right]
\]

(5.9)

Eq. 2.38 which is a simplified version of the above two equations assumes that \( \psi_1 = \pi \). But for real
lasers, the \( \psi_1 \), which is the FM/IM phase shift has been known to vary to upwards of \( \sim 1.2\pi \) depending
on amplitude and frequency of modulation \([5]\). Hence, this was calculated during characterization of the
laser diode and found out to be \( 1.3\pi \) for both the lasers. This value of \( \psi_1 \) was used here. For optically
thin transitions, the resultant 1f signal from the LIA can be approximated as \([5]\)

\[
R_{1f} = \frac{1}{2}GI_0i_0
\]

(5.10)
which is used for normalizing the 2f signal. Therefore, the 2f signal becomes independent of the average laser beam intensity and the opto-electrical gain G thus guarding against variations from beam-walking, blocking of beam by dust/dirt particles etc. It is worthwhile to note here that for tomography, projections signals need to be LOS. For an optically thin sample, individual harmonics (H1, H2, H3... etc.) are all LOS. Therefore, all linear combinations (with background signal subtracted) will also be LOS. Expressing the 2f peak height as the absolute magnitude of the vectorially added X and Y components prevents it from being a linear combination of the harmonics. For this reason the detection phase-shift is adjusted to zero and only the X-component is used for tomography.

5.3 Laser Characterization

In order to perform calibration free WMS-2f as described in Li, et al. [5], it is important that the laser be characterized beforehand. The output of the laser diode driven by laser controller given by Eq. 2.30 is reproduced once more for quick reference

\[ I_0(t) = G \bar{I}_0 [1 + i_0 \cos(\omega t + \psi_1) + i_2 \cos(2\omega t + \psi_2)] \]

and

\[ \nu(t) = \bar{\nu} + a \cos(\omega t) \]

where \( \bar{I}_0 \) is the average laser intensity at \( \nu \), \( G \) is the opto-electrical gain, \( i_0 \) is the linear (1f) and \( i_2 \) is the non-linear (2f) amplitudes of the incident intensity normalized by \( \bar{I}_0 \), \( \psi_1 \) is the linear phase FM/IM phase-shift and \( \psi_2 \) is the non-linear FM/IM phase-shift. The laser is characterized once the values \( i_0, i_2, \psi_1, \) and \( \psi_2 \) are known for a given modulation frequency and the for the range of \( \bar{I}_0 \) used in a scan.

Traditionally for fixed wavelength WMS-2f, the phase shifts are found out by modulating the laser injection current using a sinusoidal wave. The modulation not only modulates the signal intensity (which is recorded with a photodetector) but also the wavelength of the laser which follows the time-dependent spectral equation shown above. The intensity modulation is fitted with a 1f curve while its residual is fitted with a 2f curve. On the other hand, the frequency or wavelength change due to changing injection current can be determined by using a solid etalon. In short, a solid etalon is a “Fabry–Pérot interferometer” which creates transmission fringes in the wavelength domain with a resolution equal to
Figure 5.3: Finding change in wavelength of a laser by using output of an etalon

its free-spectral range (FSR). Typically it is made of a transparent solid with two highly reflecting parallel surfaces. Interference caused by multiple reflections from the surfaces creates a wavelength dependent transmission function with equi-spaced (in wavelength domain) peaks. When a laser is scanned and/or modulated using a changing injection current, its output wavelength will also change. By counting the number of fringes crossed during the time of its modulation it is therefore possible to determine the change in wavelength in the same time. Plotting the sinusoidal change in wavelength with time and the sinusoidal change in intensity with time side-by-side helps one to determine the phase-shift value for $\psi_1$ of these two sinusoids. As mentioned before, the non-linear amplitude modulation $i_2$ is then be found using the 2f-fitted curve to the residual of 1f. The corresponding phase shift is also deduced in the same manner.

This work however utilizes scanned WMS-2f and hence the procedure employed to find $i_0$, and $\psi_1$ is slightly different and is therefore described below. Figure 5.3 shows the output of a photodetector that monitors the fringe from a Si-etalon (~83mm, Lightmachinery Inc., Canada, (FSR) of ~ 0.017cm$^{-1}$) as
the laser is scanned using a sawtooth. The top figure only shows the deviation from the mean intensity due to the fringe while the bottom plots the change in output wavelength with changing injection current. Plotting the intensity output of the laser intensity for a given modulation amplitude and frequency alongside the wavelength signal from the etalon helps in determining the phase shift $\psi_1$ [5].

**Finding amplitude of modulation “a”**. The spectral modulation amplitude “a” is a key parameter that needs to be determined prior to the experiments so that results can be compared to simulation-based “look-up-tables” using HITRAN or HITEMP database. In order to employ the designed modulation amplitude that is used in the simulation the spectral tuning characteristics of the laser are hence required to be ascertained. Injection current based tuning creates changes in output wavelength of the laser that depend on both the amplitude of modulation as well as frequency of modulation. Therefore, the scanned-modulated output of the laser was passed through a solid Si-etalon with a free spectral range (FSR) of $\sim 0.017\text{cm}^{-1}$ in the 1.4$\mu$m range. The tuning characteristics (i.e. change of output wavelength – nm/mA of injection current) and amplitude of modulation “a” was found by counting the number of fringes that appear in the output in 1 complete modulation cycle.

**Finding linear modulation coefficient “$i_0$”**. Briefly, in the absence of the flame, the injection current to each laser was scanned with a frequency of 200Hz and modulated simultaneously with a frequency of 50kHz. The raw signal from the photodetector was fed into the digital “lock-in-amplifier” (LIA) (coded using LABVIEW) and the 1f output was recorded. Theoretically, if the absorbance in the spectral domain is small, then the output is given by [5]

$$R_{1f} = \frac{1}{2}G\bar{I}_0i_0$$

(5.11)

where, $R_{1f}$ (shown in Fig. 5.4a) is the 1f signal which scales with the mean detector signal $G\bar{I}_0(t)$ (shown in the inset of the same figure). In order to find $i_0$, this output was normalized by the central detector output which in turn was found by fitting a 2nd order polynomial through the raw photodetector signal and is shown in Fig. 5.4b). The normalized linear amplitude $i_0$ is found by performing a best-fit of a sinusoidal curve with the same frequency to the intensity signal and normalizing it with the average signal strength [5].

**Accounting for residual amplitude modulation (RAM)**. Equation 2.30, shows that even without the presence of the any flame, the 2nd harmonic output from the lock-in-amplifier will be non-zero due to
Figure 5.4: Measurement of laser characteristics - (a) 1f signal from LIA and raw signal (b) linear modulation coefficient (i₀)

non-linear amplitude term i.e. \(i_2 \cos(2\omega t + \psi_2)\). This is also known as residual amplitude modulation or RAM [15, 5]. RAM effects need to be vectorially subtracted when working with large modulation depths. In our work modulation depths were small (0.13 cm\(^{-1}\) for the transition at 1442.67 nm and 0.1 cm\(^{-1}\) for the transition at 1391.67 nm). For the laser emitting at 1391.67 nm, this term is negligible compared to the 2f signal values even at the largest flame radius (where only absorption from room-temperature water vapor is present). However, it is not the same for the laser at 1442.67 nm. It should be noted here that, this laser has negligible absorption at room-temperature (i.e. outside the flame) and hence the 2f signal for this laser should ideally go to zero at the edge of the flame. Instead the presence of RAM, though small, contributes significantly to the resultant 2f signal for the last 2-3 locations inside the flame. Hence, the RAM for this laser was recorded and vector subtracted (refer Eq. 5.4 through 5.9) from the measured signals.

5.4 Optical Setup, Automation and Data Acquisition

Optical setup. The experimental setup (shown in Fig. 5.5) for 1D tomography consists of an arrangement for scanning the McKenna flame by a single laser beam moving parallel to itself. The attenuated intensity of the beam is recorded by a fixed photo-detector (PDA-10CS, Thorlabs, USA). This is achieved in the following manner – the laser beam emitted from the diode (NLK1E5C1TA, NTT Electronics, Japan), mounted in a \(\phi - 9\) mm TO-can (LDM-4405, ILX Lightwave, USA) mounting fixture, is
collimated using an aspheric lens and shaped using an iris. The output of the second laser is processed similarly and the two beams are combined into single collinear beam using an appropriate pellicle beam splitter (CM1-BP145B3, Thorlabs, USA). One of the arms of this beam splitter is directed into the solid Si-etalon mentioned before and its output is recorded using a photodetector enabling close monitoring of the wavelength. The other beam-arm is then directed vertically downwards using a periscope arrangement so that it becomes co-axial with the vertical axis of a rotational stage (RGV-100 BL, Newport Corp., USA). A plane mirror mounted at 45° to the axis is used to reflect the incoming vertical beam along a horizontal plane while the rotational stage is used to rotate this reflected beam in the horizontal plane. The center of the angled mirror is coincident with the focus of an off-axis parabolic mirror. Therefore, as the plane mirror is rotated by the rotational stage, the reflected ray from the parabolic mirror always moves parallel to itself thus scanning the surface of the burner. On the other side of the burner, another parabolic mirror collects all the reflected rays and directs them into the sensitive area of the photode-
tector placed at the focus of the second parabolic mirror. The laser emission wavelength and intensity is controlled by a laser controller (ILX Lightwave, LDC-3908) which is capable of controlling both the injection current and temperature of the diode. The temperature control is used to set the central emission wavelength of the laser while the injection current is scanned and modulated using a programmed function generator (CG4340, Gagecard). The two lasers were multiplexed in the time domain i.e. when laser 1 was active laser 2 was waiting and vice-versa. This firing strategy is graphically shown in Fig. 5.6). The flame was then scanned by laser beams that move parallel to itself with a spatial resolution of 1mm. The size of the beam spot was of the order of thickness of the graduation on a standard meter scale i.e. less than 0.5 mm.

**Automation.** The movement of the rotational stage is controlled by the controller and driver cards (XPS-C4 and XPS-DRV02, Newport Corp., USA ) which were programmed using LabVIEW as well. The stage has a maximum speed of 720 degrees/sec and the burner which is 37.5 mm (including the shroud flow) in radius subtends ~ 8 degrees at the focus of the parabolic mirror. Therefore, with a rapid enough data acquisition system, one laser sweep across the burner can be completed inside ~\(\frac{1}{10}\)th of a second. However, instead of running the stage at full speed, we currently move the stage to a pre-determined set of angles (dictated by the ray spacing) and collect beam attenuation data at those points.
100 sets of 10 sawtooth scans (5 of each laser) are recorded at each location.

The flame used here is a methane-air flame flow rates of which are controlled by mass flow-controllers (Model# 32907-71 and 32907-73, Cole-Parmer, USA). The flame is guarded by a shroud flow of nitrogen the linear velocity of which was matched with the linear velocity of the cold-flow of the air-fuel mixture in order to minimize shear at the edge of the flame. The nitrogen flow was also controlled by a flow controller (#32907-75) and controlled from a computer. All flow-controllers were programmed using LabVIEW. The controllers are capable of limiting the flow rates to ±2% of the set-point value. An equivalence ratio of 0.7 and a mixture flow velocity of 20 cm/sec was used for the experiments. In order to cool the burner matrix, water was pumped through the cooling coils at a rate of 800 ml/min. Input temperature of the water was held constant at 4°C by using a mixture of ice and water held in an enclosed sump.

**Data Acquisition.** The function generator uses a 200Hz scanning ramp super-imposed with a 50kHz sinusoidal modulation to create the harmonics of the absorption signal which is recorded by the photodetector and sampled by a digital oscilloscope (NI PCI-5105 digitizer) at a high enough sampling rate using a LabVIEW NI-scope program. The same function generator, while controlling the injection current of the lasers, also generate a square wave that is synchronous with the sawtooth scans of the laser injection current and has the same time period as well. The square wave output of the function generator and the output of the photodetector are fed into 2 different channels of the digital oscilloscope. Once the beam moves into position, the information is sent to the digitizer which has been monitoring the square wave signal level continuously. The digitizer then uses the falling edge of the next available square wave as trigger to start acquiring samples from the photodetector output. This makes sure that data acquisition always occur at the very beginning of the sawtooth thereby providing a fixed reference point to plot 2f signal level versus time (or wavelength). A chunk of 10 sawtooths are captured and processed using a digital lock-in-amplifier (programmed in LabVIEW) that extracts the different harmonics of the modulated signal (1f, 2f and 3f) and writes them to a binary file.

**5.5 Determination of Temperature and Concentration**

Traditionally, WMS-2f requires modulation amplitudes of the lasers be optimized based on the temperature and concentration under investigation [30, 29]. When the modulation index “m” is around 2.2 i.e.
when the amplitude of modulation is 2.2 times the half-width of the spectral transition, the laser is said be optimized. Near $m = 2.2$, the 2f signal strength is maximized and the 2f ratio shows little variation with small changes ($\pm 10\%$) concentration and or pressure as shown in Fig. 2.6. In other words, the strength of the second harmonic, $H_2$, in Eq. 2.36 can be considered a linear function the concentration $X$, because the lineshape function “$\phi$” term inside the integral will be almost constant thereby making the ratio of 2f heights independent of concentration. Working within this framework, one can then use the LUT (look-up-table) of ratio vs temperature simulated for the ball-park concentration and find a temperature with minimal error. However, in the context of tomography, it should be remembered that concentration is neither constant over space nor varying within just $\pm 10\%$ of a mean value. In fact, for a McKenna flame concentrations are expected to vary between 0 (near the edge of the flame) to about 14 mole percent at the center of the flame. An error in selecting the correct concentration LUT will obviously create large errors in temperature (shown in Fig. 5.7). Hence, away from the concentrations for which the modulation amplitude can be considered nearly optimized, the $H_2$ is non-linear function of concentration and ratio of 2f heights is dependent on concentration.
Figure 5.8: Iterative 2 line temperature and concentration determination (Red denotes starting guess higher than final concentration, blue denotes starting guess lower than final concentration).

5.5.1 The Iterative Approach

To circumvent the problem mentioned above, an iterative method of temperature determination was employed. This scheme is shown in the flowchart Fig. 5.9. In short, we start by assuming a concentration (0.01 mole fraction, by default) and using the LUT for ratio of 2f vs temperature find the temperature which is then used to find the concentration. Implicitly, this assumes than at every temperature step the H2 signal is proportional to concentration thereby incurring an error in both concentration and temperature determination. This error is then reduced by continuing the iteration further till a suitable cutoff criteria is satisfied ( when increment of concentration is less than 1% of the increment in previous iteration). As an example, we used simulated spectral data 2f height for a temperature of 1080 K and 7 mole percent. In the absence of any measurement error, the converged solution matched the temperature and concentration exactly. Then 1% measurement error was introduced in one of the spectral line data and the converged solution was found to be 1090 K and 7.1 mole percent. The march of temperature and concentration for every iteration is shown in Fig. 5.8. The figure also shows that the initial guesses (viz. 9 and 5 mole percent, which is far from the ±10% margin of 7 mole percent ) does not affect the final outcomes.
5.5.2 The Non-linear Least Squares Approach

The other method of solving this problem would be to find the best possible values of temperature and concentration that explain the two spectral 2f height measurements. The advantage of this approach over the previous one is that it is easily extendible to more than 2 transitions if such is ever used for the purposes of increasing accuracy. To solve this we define a variable “$\Delta$” as

$$\Delta = \sqrt{\sum_{i=1}^{2} \left( \frac{\alpha_i - \beta_i}{\beta_i} \right)^2} \tag{5.12}$$

where the subscript “$i$” denotes the spectral transition (2 in this case, but can be extended to more transitions if required) $\alpha$ is the simulated value of the 2f height at a given temperature and concentration while $\beta$ is the measured value of the 2f height at that spectral transition. This is solved using a Levenberg-Marquardt scheme with temperature and concentration as free parameters. The surface $\Delta$ shown in Fig. 5.10, has one minima for a given temperature and concentration within the range of temperature and concentration we expect to work inside which represents the converged solution of the scheme.
5.6 1D Tomography Results

For 1D tomography, measurements were taken at a height of 7mm from the burner plate with an equivalence ratio $\phi = 0.7$, and a linear velocity of 20 cm/sec. The justification for using the such a low value of $\phi$ stems from the fact that higher equivalence ratios produce significantly higher temperatures. Sensitivity for temperature measurements (and therefore concentration too) drops with temperature (Fig. 5.1). Since the measurement accuracies are limited by the sensitivity of the available pair of spectral transitions, efforts were made to produce a flame with a low temperature i.e. low $\phi$.

**Individual Line Reconstructions.** The following figures show reconstructions for individual spectroscopic lines i.e. the 1f normalized 2f values at different radial locations. Abel 3 point (ATP) was used for the de-convolution while the regularization parameter $\lambda$, was taken as 1 following the L-curve criterion [73]. The reconstructions were created from 100 sets of scans – each set containing 5 sawtooth scans of each spectroscopic line using time-domain-multiplexing. The projection data for the two spectroscopic transitions (i.e. the 1f normalized 2f LOS measurements) are shown in Fig. 5.11. The burner radius is 30 mm but repeated measurements at $r = 30$mm, with laser 1 showed a non-zero 2f signal value whereas measurements at $r = 31$mm showed values that were of the order of the residual amplitude modulation (RAM). Therefore, the $r = 31$mm was taken as the edge of the flame and the RAM values there were
vector-decomposed and subtracted from other measurements. This penetration of the flame into the shroud flow ($N_2$) is clearly seen from Fig. 5.12a and b. As for the second spectroscopic transition, LOS values for the flame were determined by subtracting the outside $2f$ signal (at $r=31$mm, scaled and corrected for the size of the burner at each cord location). The $2f$ signals scale linearly with pathlength as long as the assumption $1 - \exp(-\alpha) \approx \alpha$ holds good, which it does for $\alpha < 0.1$. Since the maximum absorbance for the second line was below 0.1, this method is expected to work well. The local radial values of the distribution shown here are scaled for 1cm path length instead of 1mm to match the simulation “look-up-tables” which were created for 1 cm.

**Look-up-table simulation.** A “master” look-up-table for $2f$-peak heights near the two transitions w.r.t. temperature was created for different concentrations (varying between 0.01 to 0.16 mole fraction of water vapor). In order to do this, firstly a full scale simulation for total spectral absorbances (i.e. $\alpha(\nu)$ versus $\nu$) was carried out near the spectral regions of interest. Parameters like line-strength, transition wavenumbers, self-collisional coefficients etc. were taken from the HITEMP database [20, 57]. Transitions with line-strengths less than $1e-34$ cm$^{-1}$/molecule.cm$^{-2}$ were neglected. Using the data from HITEMP, absorbance curves were simulated for different temperature and concentrations. Subsequently, $1f$ normalized $2f$ peak heights in Eq. 2.41 were simulated by taking a fast-Fourier transform (FFT) of the absorbance line shape around the transition line-centers where the proper experimental values of modulation amplitude “$a$”, linear modulation amplitude coefficient “$i_0$” and phase shift “$\psi_1$” was substituted.
This “look-up-table” was used to determine temperature and concentration of each radial point inside the flame with the measured values of the 2f heights as inputs.

**Temperature and Concentration** Using the values of individual line reconstruction and following the methodology explained in section 5.5.2, the temperature and concentrations of the flame was reconstructed as well. Fig 5.13 shows the results for temperature and concentration reconstruction. The uncertainty values were based on measurement uncertainty and propagation of uncertainty in Abel inversion. In Abel’s inversion, uncertainties tend to grow towards the center which is clearly seen from the results. Near the center the uncertainties are around ±70 K with a temperature of around 1670 K. Concentration values near the center are around 0.135 mole fractions of water vapor with an uncertainty of ±0.0077 mole fraction. Both temperature and concentration remain constant for nearly 10mm away from the radius beyond which gradients become perceivable. For r>23mm, the values for temperature and concentration drops drastically.

**Validation of results.** The species concentration values near the center of the flame are expected to be close to equilibrium concentrations regardless of the other parameters like matrix material of the burner, cooling water velocity and temperature, gas flow rates etc. Previous work by Villareal et al. [53] (who used DA spectroscopy and Abel inversion on a McKenna burner, matrix material or other parameters unreported) as well as work done by the DLR institute, Germany (coherent anti-Stokes Raman spec-
troscopy or CARS on a bronze matrix McKenna burner) [67] show that concentration values of species (CO₂ and H₂O) are very near equilibrium values near the center of the flame. Our results also show the same, (reconstructions near the center are within 1.5% of equilibrium values) which validate these measurements.

In contrast to concentration values, the flame temperature values depend significantly on a number of parameters. In a burner stabilized flame like a McKenna burner, the flame temperature is always lower than adiabatic flame temperature (T_{ad} = 1838K in this case) due to heat transfer from the flame zone to the burner plate. Heat transfer depends on a number of parameters like burner matrix material, flame stabilization height, flow velocity, burning velocity, cooling water temperature and flow rates. Additionally, burner to burner temperature differences of the order of 25K has been noticed for identical parameters [67]. Also, it was shown by Villareal et. al [53] that at high temperatures thermocouple results are unreliable because of high uncertainties in radiation correction. Migliorini et al. [90] showed that even with all other parameters same, the center temperatures of the flame is ~70K lower for a stainless steel matrix burner at a flame temperature of ~ 1300K. As such, a direct comparison of temperature results with other existing studies is not justified. However, it must be noted that for a given temperature and concentration, spectral absorbance and 2f heights are single valued. Therefore, validation of concentration implicitly validates temperatures as well.
(a) Temperature reconstruction using Tikhonov regularized inversion

(b) Concentration reconstruction using Tikhonov regularized inversion

Figure 5.14: 2D temperature and concentration reconstruction (5 views, 60 rpm)

(a) Comparison of 2D temperature reconstruction with 1D result.
(b) Comparison of 2D concentration reconstruction with 1D result.

Figure 5.15: Comparison of 2D reconstruction and Abel results at burner mid-section
5.7 2D Tomography Results

For 2D tomography, the area around the burner was discretized into a square domain of $62 \times 62$ pixels each with sides 1mm length (3844 pixels). The domain circumscribes the burner completely. A data set from 1D tomography for both the transitions were copied for 5 views that subtended equal angles ($36^\circ$) between themselves. Each view was assumed to consist of 60 equi-spaced rays and Tikhonov regularized inversion was performed with $\lambda$ value of 4 (following procedures mentioned in section 3.4.2). Deconvolution of spectral values was smoothed using a median filter of size $5 \times 5$ pixels before temperature and concentration reconstruction and the same was applied to the final temperature and concentration values to mitigate TDLAS artifacts. Resulting reconstructions are shown in Fig. 5.14a and b. It can be seen that the overall outer edge of the flame is well approximated by the reconstructions except some errors from the low signal regions. Minor artifacts are seen near the $x=\pm10-20$mm where the circular shape gives way to a more polygonal feature accompanied by a steep change in temperature and concentration. The same is visible in the cross-section cut across the center of the flame and compared to the 1D results (Fig. 5.15). Overall, both concentrations and temperatures near the center are well approximated. However, the concentration gradient near the center is non-zero unlike in 1D case. On the other hand, concentration gradients near the edge follow the 1D case better than the temperature comparisons. These reconstructions show significant potential for a true 2D tomographic setup.
Chapter 6
Conclusions and Outlook

Application of TDLAS techniques to non-homogenous temperature and concentration domains is an area where very few studies exist. Even fewer exist with WMS-2f that use only 2-spectral lines. This work focuses on addressing this area by exploring the application of tomography in conjunction with line-of-sight WMS-2f TDLAS. For 1D tomography, it was shown that a high resolution yet, smooth solution could be achieved using Tikhonov regularized Abel inversion instead of traditional Abel inversion with coarse grids. For 2D tomography Tikhonov regularized inversion yielded solutions that match reasonably well with 1D solutions and did not require a good initial guess (as is required for iterative techniques like MART). The study also showed that the WMS-2f technique which traditionally requires its modulation amplitudes to be optimized based on known concentration and range of temperatures can be extended to non-optimal modulation depths with completely unknown concentrations. Other specific contributions of this study are listed below.

Tomography. For 1D tomography, instead of using traditional Abel’s inversion with large ray spacings, averaging and Tikhonov regularization with closely spaced rays were used. While closely packed rays provided greater resolution of the domain, averaging and Tikhonov regularization helped in stabilizing the solution against the detrimental effects of noise in projections exacerbated by the proximity of the rays. Analysis of the two kind of errors involved in Abel’s inversion – regularization error and perturbation error, showed that of the different existing algorithms, ATP is least prone to noise amplification for a given order of accuracy. Numerical results from design-of-experiments with flexible ray spacings showed that equi-spaced rays are not ideal from the point of view of optimizing perturbation error. On the other hand, attempt to optimize ray spacings based only on perturbation error, introduces significant errors in accuracy. In contrast to 1D tomography, which is mathematically an ill-conditioned full rank problem, 2D tomography is a rank-deficient problem. In 2D tomography, the regularization error dominated the perturbation error. Tikhonov regularization helps in 2D tomography by adding a-priors like “smoothness” and “non-negativity” to the solution.
**Artifacts.** It was found that artifacts in 1D and 2D tomography stem from different sources. In 1D deconvolution, artifacts are mainly due to amplification of the measurement noise in the final solution. Tomographic TDLAS using 2-line thermometry contribute to artifacts mainly through uncertainties of spectral data which may be large in low-signal regions. On the other hand, in 2D de-convolution, seemingly negligible artifacts from individual spectral-line reconstruction in low signal areas maybe be magnified by tomographic TDLAS even in the absence of any measurement noise.

**Experimental Methods.** Only one laser-detector pair along with a rotating mirror was successfully used to scan across a stationary premixed laminar flat flame with rotationally symmetric distribution to record ray-path integrated absorption spectroscopy data. Instead of a large number of spectral transitions, only two transitions were used to reconstruct the spatially varying features of temperature and concentration of the flame. The method of choice was calibration-free wavelength modulation spectroscopy with second harmonic detection (WMS-2f). Traditionally, for WMS-2f, it is implicitly assumed that the concentration is uniform and known to within $\pm 10\%$ of the mean value. Based on this, the modulation index is generally optimized to near 2.2 so that minor variations in concentration or pressure do not affect the final outcome significantly. In contrast, this study used WMS-2f where both temperature and concentrations were unknown and varied widely over the whole domain (water vapor concentrations vary from nearly zero near the edge of the flame to 14 mole percent at the center). Hence, the modulation indices for the two lasers were “non-optimal” for this case. It was shown that for such a case, both an iterative procedure or a least-squared approach was can be employed to determine temperature and concentrations. Final concentration results matched well with expected equilibrium values near the center of the flame (where diffusion effects are minimal). Also, since for a given temperature and concentration, 2f spectroscopic signals are single valued, correct concentration results implicitly validate temperature results. Taking insights from simulation work done earlier during this study, the set of experimental data was extrapolated in 2D (i.e. copied for different views) and 5 views with 60 rays per view were used to reconstruct the temperature and concentration of the domain without the use of the axial symmetry assumption for the flame property distribution. The 2D results too match reasonably well with the expected profile.
Outlook and recommendations. Based on the above observations it can be concluded that further improved results maybe obtained using some additional spectral lines. In case of 1D tomography, higher sensitivity line-pairs as well as additional spectral lines can reduce uncertainties of measurements. In case of 2D tomography, the same strategy can be used to reduce artifacts in reconstruction. The current solution method uses Tikhonov regularization on individual spectral data distributions and combine them to determine temperature and concentration. A solution method that takes into account not only the smoothness and non-negativity of the spectral distribution but also smoothness and non-negativity of the final temperature and concentration, possibly in an iterative manner, may eliminate the need of any median filtering and reduce tomographic TDLAS artifacts in the final reconstruction significantly. Also, a spectral line that probes a different species e.g. CO$_2$, may be used to obtain additional smoothness and accuracy information.

Finally, the existing experimental setup and automation can be modified to build a true 2D tomographic experimental setup. Five rotating mirrors scanning the flame, paired with five parabolic mirrors can be used to gather information from different directions. In order to reduce alignment optics which contribute significantly to disturbances in the data (from back reflections, temperature variation of optical components etc.), optical-isolator included butterfly-mounted fiber-optic lasers can be multiplexed to produce a single collimated beam. This beam can be split using fiber splitters which can lead individual beams to the mirrors mounted on rotating tables. The positions of the mirrors and the angles of the rotation of the beams can be optimized using the D-optimality routine described in this work so as to produce reconstructions with as few artifacts as possible. Thus limited view tomography with ~ 5 views is feasible.
References


Appendix
Abel’s Inversion : Derivations

The derivation roughly follows that of Dasch’s Abel Three-Point (ATP) algorithm, albeit non-uniform ray spacing and a discretization scheme based on four grid points are used. Discretizing the Abel transform (Eq. 3.1) using rays positioned at radii \( r_i \), the Abel transform for the j-th ray positioned at \( r_j \) is given as

\[
P(r_j) = 2 \int_{r_{i=j}}^{r_{i=j}+\Delta r_i/2} \frac{F(r')r'}{(r'^2-r_j^2)^{1/2}} \, dr' + ..
\]

\[
.. + 2 \sum_{i=j}^{m-1} \int_{r_{i-\Delta r_i/2}}^{r_{i+\Delta r_i/2}} \frac{F(r')r'}{(r'^2-r_j^2)^{1/2}} \, dr'
\]

\[
.. + \left[ \ell(r_j) - 2 \left( \frac{r_m^2 - r_j^2}{2} \right)^{1/2} \right] F_{\infty}
\]

where the truncation after point \( r_m \) implies that \( F(\ell > r_m) = F_{\infty} \) and the length of the ray is assumed to be \( \ell(r_j) \).

Introducing \( \Delta r_i \equiv r_{i+1} - r_i \) and \( \delta \equiv (r' - r_i) / \Delta r_i \), the function value \( F(r') \) is rewritten as

\[
F(r') = (1 - \delta)F(r_i + \delta \Delta r_i) + \delta F(r_{i+1} - (1 - \delta) \Delta r_i)
\]  (6.1)

The derivatives are approximated in the vicinity of \( r_i \) and \( r_{i+1} \) by the linearized expressions

\[
F(r_i + \delta \Delta r_i) \approx F_i + F'_i \delta \Delta r_i
\]

\[
F(r_{i+1} - (1 - \delta) \Delta r_i) \approx F_{i+1} - F'_{i+1} (1 - \delta) \Delta r_i
\]

respectively, using locally evaluated \( F_k = F_k(r_k) \) and \( F'_k = F'_k(r_k) \). Through these local approximations, Eq. 6.1 takes the form of a second-order spline function. Defining \( d_k \equiv r_k / \Delta r_i \), the integrals are rewritten as

\[
P(r_j) = \sum_{i=j}^{m} \int_{0}^{1} \frac{1}{r_i} \left[ f_i^{(0)}(\delta) + f_i^{(1)}(\delta) \delta + f_i^{(2)}(\delta) \delta^2 + f_i^{(3)}(\delta) \delta^3 \right] \, d\delta
\]  (6.2)
where \( f^{(\ell)}_i \) are constants defined as

\[
\begin{align*}
 f^{(0)}_i &= 2\Delta r_i d_{ii} F_i, \\
 f^{(1)}_i &= 2\Delta r_i \left[ (1 - d_{ii}) F_i + d_{ii} F_{i+1} + d_{ii} (F'_i - F'_{i+1}) \Delta r_i \right], \\
 f^{(2)}_i &= 2\Delta r_i \left[ \Delta r_i (d_{ii} - 1) (F'_{i+1} - F'_i) - F_i + F_{i+1} \right], \\
 f^{(3)}_i &= 2\Delta r_i^2 (F'_{i+1} - F'_i)
\end{align*}
\]

Based on a known ray spacing, all integrals can be solved a priori, and the Abel transform becomes

\[
P(r_j) = \sum_{i=j}^{m-1} \left[ f^{(0)}_i I^{(0)}_{ij} + f^{(1)}_i I^{(1)}_{ij} + f^{(2)}_i I^{(2)}_{ij} + f^{(3)}_i I^{(3)}_{ij} \right]
\]

(6.3)

where \( I^{(n)}_{ij} \) are evaluated as

\[
I^{(n)}_{ij} \equiv \int_0^1 \frac{\delta^n}{\left( (d_{ii} + \delta)^2 - d_{jj}^2 \right)^{1/2}} d\delta
\]

Thus far, there have been no assumptions for the evaluation of the constants \( p^{(n)}_i \), which are formed from local first and second derivatives of the projections. Using three-point finite difference expressions in points \( r_i \) and \( r_{i+1} \), it follows that

\[
\begin{align*}
 F_i &= a_i F_i^T = \begin{bmatrix} 0 & 1 & 0 & 0 \end{bmatrix} F_i^T \\
 F'_i &= b_i F_i^T = \begin{bmatrix} a_i & a_i^\circ & a_i^+ & 0 \end{bmatrix} F_i^T \\
 F_{i+1} &= c_i F_i^T = \begin{bmatrix} 0 & 0 & 1 & 0 \end{bmatrix} F_i^T \\
 F'_{i+1} &= d_i F_i^T = \begin{bmatrix} 0 & a_{i+1} & a_{i+1}^\circ & a_{i+1}^+ \end{bmatrix} F_i^T
\end{align*}
\]

where \( F_i \) is defined as

\[
F_i \equiv \begin{bmatrix} F_{i-1} & F_i & F_{i+1} & F_{i+2} \end{bmatrix}
\]
and the coefficients $a_k^{-/+}$ and $b_k^{-/+}$ are easily obtained from a Taylor expansion as

$$
\begin{align*}
\alpha_k^{-} &= \frac{-\Delta r_k}{\Delta r_{k-1}(\Delta r_{k-1} + \Delta r_k)} \\
\alpha_k^{o} &= \frac{\Delta r_k - \Delta r_{k-1}}{\Delta r_{k-1} \Delta r_k} \\
\alpha_k^{+} &= \frac{\Delta r_{k-1}}{\Delta r_k (\Delta r_{k-1} + \Delta r_k)}
\end{align*}
$$

except for $i = 0$ and $i = m - 1$, where simpler two-point finite differences are used.
**Vita**

Avishek Guha was born and raised in Calcutta, India. After finishing higher secondary education from South Point High School, he went to Jadavpur University, Calcutta, where he received his baccalaureate degree in mechanical engineering in 2003. Between 2003 and 2007 he worked in the automotive manufacturing industry in India. In the fall of 2007 he joined the master’s program in mechanical engineering at LSU and received his masters of science degree in the fall of 2009. He started his doctoral program at LSU in the spring of 2010 under the guidance of Dr. Ingmar Schoegl and will graduate in May of 2014. After graduation he expects to join Air Products and Chemicals, Inc. as a research engineer.