Characterization and Improvements of Filtered Rayleigh Scattering Diagnostics

THESIS

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Abstract

Described in this thesis is an analysis of the fundamental characteristics and typical limitations associated with the filtered Rayleigh scattering (FRS) measurement technique. Particular emphasis will be placed on the applicability of the technique toward obtaining gas-phase information through Rayleigh scattering in flowfields traditionally dominated by unwanted light scattering. This includes measurements conducted simultaneously with particle imaging velocimetry or in multi-phase flows such as spray environments and sooting flames, where Mie/Tyndall scattering overwhelms gas-phase Rayleigh scattering or experiments in enclosed environments, with large amounts of residual laser scattering.

A preliminary overview of the FRS technique will be presented, along with previous work utilizing the technique in a variety of applications including flow visualization in high speed and compressible flows, temperature, pressure and number density measurements in flames and turbulent flows, as well as simultaneous FRS and particle imaging velocimetry in flames. Experimental limitations arising in these experiments will be discussed.

Essential to the filtered Rayleigh scattering technique is a thorough understanding of the spectral and thermodynamic properties of the atomic or molecular filter used to transmit portions of the gas-phase Rayleigh scattering signal to a detector while
suppressing the interfering Mie/Tyndall/surface scattering. As such, a model of the transmission (or conversely, the absorption) characteristics of the molecular iodine cell utilized in this work will be presented. A parametric study of the effects of cell temperature and pressure will be described; with an emphasis on the impact of varying these parameters on the maximum scattering rejection attainable by the filter cell. Subsequently, experimental analysis of the suppression capabilities of our particular I₂ cell will be presented. Of particular note are the fundamental differences in cell transmission observed when using a narrow linewidth continuous wavelength (cw) laser source, as compared to an injection-seeded, Q-switched, pulsed laser source. It was determined that the disparities in filter cell transmission arising from these two laser sources is fundamentally related to the spectral purity of the pulsed laser, a measure of the proportion of energy contained in a single spectral mode to the total energy of the illuminating laser source.

Results obtained both experimentally and through the use of a simple model will be presented highlighting the functional dependence of the maximum absorption of the iodine filter cell on the spectral purity of the laser source. It also will be shown that there is an exponential relationship between the seed laser power (when using an injection seeded Q-switched, pulsed laser source) and the spectral purity of the laser output. The results of this study subsequently are related back to previously-observed limitations of the technique available within the literature.

One potential methodology to increase laser spectral purity investigated in this thesis involves the insertion of a Fabry-Perot etalon into the external optical path of an
injection-seeded Nd:YAG laser. A description of the transmission properties of an etalon (or similarly a Fabry Perot interferometer) will be given. Particular emphasis is placed on how the etalon is used to effectively increase the spectral purity of the transmitted laser pulse. Experimental results demonstrate improvements in the laser spectral purity greater than one order of magnitude when using the etalon. In terms of FRS measurements, scattering rejection was increased by more than two orders of magnitude when using the etalon.

Finally, initial validation studies, where gas-phase information is extracted from droplet-laden (non-evaporating) turbulent jets of propane and air, is presented. Comparisons of suppression capabilities with and without the etalon in the optical path are shown, highlighting the gains resulting from implementation of the etalon. Finally a demonstration of the technique is presented within an evaporating spray jet of acetone, highlighting the potential of the FRS technique for extracting gas-phase information within turbulent spray flows.
Dedication

This document is dedicated to my family and friends.
Acknowledgments

I would like to thank my friends and family for their continued encouragement. I thank Dr. Naibo Jiang for his early guidance, hard work, and genial attitude. I thank the students of NETL and TCRL for their unwavering support and for helpful conversations. I thank my wife, Jessica Patton, for being my inspiration.
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Chapter 1: Overview of the Filtered Rayleigh Scattering Technique

Laser-based diagnostics have allowed for multi-dimensional, non-intrusive measurements in a variety of flowfields of interest in turbulence and combustion. While advancements in terms of laser energy and camera sensors have led to increasing resolution and accuracy, there exist a number of situations or environments where traditional laser-based diagnostics remain challenging such as a sooting flame, a spray, or an experimental apparatus with limited optical access where laser scatter may be unavoidable.

Of particular interest is the ability to obtain quantitative information within these difficult environments, while maintaining the benefits associated with a two-dimensional laser measurement techniques. Filtered Rayleigh Scattering (FRS) provides a unique methodology to measure multiple flow properties utilizing a single laser source and detector. Briefly, filtered Rayleigh Scattering is a variation of the traditional Rayleigh scattering technique, in which an optically-thick molecular or atomic filter (placed in front of a detector), is used in conjunction with a narrow linewidth laser source to collect a portion of the Doppler-broadened molecular (gas phase) Rayleigh (-Brillouin) scattering while blocking unwanted scattering of the laser light [1-5].
A graphical representation of the FRS approach is shown in Fig. 1. When a narrow-linewidth laser beam passes through a multi-phase medium\(^1\) (e.g., gas/solid, gas/liquid), the scattered light is comprised mainly of Mie/Tyndall scattering (from solid particles or liquid droplets), Rayleigh and Raman scattering (from gas-phase atoms or molecules), and stray light from surface (e.g., wall or window) scattering. The spectrum of each individual scattering process is broadened and shifted based on the motion of the scattering medium. The scattering from the surfaces in the system (“stray light”) has the same frequency distribution as the laser. Mie/Tyndall scattering from particles and droplets exhibits essentially the same frequency distribution as the laser with a small spectral shift which is a function of velocity.

Rotational Raman scattering is the result of the inelastic scattering process of molecules in which the scattered photons gain or lose energy during the interaction. The change in energy is represented by a change in frequency of the irradiating photon away from the incident laser frequency. However, this process is inherently weak and thus the scattered signal is \(10^2\text{-}10^3\) times weaker than the spontaneous Rayleigh scattering and can be neglected in the context of this work.

Rayleigh scattering from gas-phase molecules is broadened by molecular motion, which is a combination of thermal and acoustic motion. As noted in Fig. 1, this broadening extends an appreciable, and experimentally resolvable, distance away from the illuminating laser frequency. The total intensity of the scattered light is proportional to the number density of the gas and its individual scattering cross section; the spectral

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\(^1\) Plasma/neutral gas mixtures are not considered. If considered, Thomson scattering from electrons also would contribute to the scattering spectrum.
lineshape is proportional to both temperature, pressure and a function of gas properties, and the width of the spectrum can be related to the temperature and the molecular weight of the gas of interest. [6,7].

Figure 1. Filtered Rayleigh scattering concept.

If the laser source is sufficiently spectrally-narrow and is tuned such that its frequency corresponds to an absorption transition of the atomic/molecular species of the filter, then a portion of the unwanted scattering signal (e.g., Mie/Tyndall or surface scattering) will be absorbed as depicted in Fig. 1. The exact level of scattering suppression due to absorption is highly dependent on the filter characteristics as discussed below. However, as can be seen in Fig. 1, the scattered light from gas-phase molecules (Rayleigh-Brillouin Scattering) is broadened and a significant portion of this
signal will transmit to the detector and can recovered without interference from the more intense Mie/Tyndall or surface scattering.

The signal detected at each pixel of the camera is a function of the flow velocity, wavelength (or frequency, $\nu$), temperature ($T$), observation angle ($\theta$), gas composition, and pressure ($P$), due to the overlap between the scattered light lineshape and the narrowband absorption filter. This differs from the case where the Rayleigh scattering is imaged directly, where no spectral information is retained and the signal is directly proportional to number density and mixture-averaged scattering cross-section of the flow. The distribution of the scattered intensity can be written as

$$I(\nu) = AI_i n \sum_{i=1}^{N} X_i \sigma_i r_i (P,T,W,\theta,\nu)$$  \hspace{1cm} (1)$$

where $r_i$ is the convolution of the laser lineshape and the Rayleigh-Brillouin scattering profile of the $i^{th}$ species calculated from kinetic models such as the S6 model from Tenti et al. [7-10]. The total intensity incident upon a camera pixel is the convolution of the spectral intensity $I(\nu)$ and the filter transmission, $T(\nu)$

$$S(\nu) = \int_{-\infty}^{\infty} I(\nu) T(\nu) d\nu .$$ \hspace{1cm} (2)$$

$S(\nu)$ can be normalized by the scattering from a reference quantity at known temperature and pressure such as ambient air to account for the constant $A$ and the incident laser intensity $I_o$. 

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Rayleigh Scattering Properties

The accuracy of any quantitative measurements made utilizing the FRS approach will in part be determined by the accuracy to which the convolution of the laser lineshape and the Rayleigh-Brillouin scattering profile \( r_i \), can be experimentally determined or accurately modeled. Classically, Rayleigh scattering is approximated as the elastic scattering of light generated when molecules or small particles are forced to oscillate by an applied electromagnetic field. As a generalization, particles are said to be in the “Rayleigh regime” when the particle diameter is less than 1/10 the wavelength of the incident light. Therefore, if using a frequency-doubled Nd:YAG operating at a nominal value of 532nm, particles with diameters smaller than ~53 nm will scatter light in a manner appropriately approximated as Rayleigh scattering, which is the small-particle limit to the more general Lorenz-Mie-Debye scattering theory [11]. It is often convenient to define a measure of the amount of scattered energy as a ratio of incident energy for a given solid angle. This is commonly known as the differential scattering cross section, denoted by \( d\sigma/d\Omega \).

For an isotropic gas this expression is given as

\[
\frac{d\sigma}{d\Omega} = \frac{4\pi^2(n-1)^2}{N^2\lambda^4} \sin^2 \varphi \tag{3}
\]

where \( n \) is the index of refraction, \( N \) the number density, \( \lambda \) the wavelength of the incident light, and \( \varphi \) the scattering angle with respect to the incident radiation. It is important to note the \( \lambda^{-4} \) dependence of the scattered light. It is for this reason that some Rayleigh scattering measurements, including Filtered Rayleigh scattering, are performed in the ultraviolet (UV) portion of the spectrum. However, due to limitations in spectral
responses of imaging equipment in the UV region and the generally lower levels of laser pulse energy generated in the UV, the gains afforded by the $\lambda^{-4}$ dependence are not necessarily realized as increases in signal or signal-to-noise levels. While the expression of the differential scattering cross section becomes appreciably more complicated for anisotropic gasses, it retains its proportionality to number density, and its dependence on $\lambda^{-4}$[6].

For traditional laser Rayleigh scattering, the spectral lineshape of the scattered light is of little consequence. However, with an application to the filtered Rayleigh scattering diagnostic, it is this lineshape that contains information pertaining to the temperature and pressure of the scattering media. The profile of the scattered light is a reflection of the translational molecular motion of the scattering molecule, giving rise to small shifts in the frequency of the scattered light. The Doppler shift associated with this motion is given by

$$\Delta v_D = \frac{1}{2\pi} V \cdot \kappa$$

(4)

where $V$ is the velocity vector of the molecule or particle and $\kappa$, defined as the scattering wave vector is given by

$$\kappa = \frac{2\pi}{\lambda} \left( \hat{k}_s - \hat{k}_i \right)$$

(5)

with a magnitude of

$$\kappa = \frac{4\pi}{\lambda} \sin \frac{\theta}{2}$$

(6)

In Eq. (6), $\theta$ is the angle between the incident and scattered wave vectors, $\hat{k}_i$ and $\hat{k}_s$ respectively.
If the gas density is low, or if the temperature is sufficiently high, the spectral distribution of the scattered light is dominated by Doppler broadening and takes on a Gaussian distribution given by [11]

\[
g(\theta, T, \nu) = \frac{2}{\Delta \nu_T} \sqrt{\frac{\ln 2}{\pi}} \exp \left[ -4\ln 2 \left( \frac{\nu}{\Delta \nu_T} \right)^2 \right] \quad (7)
\]

where \( T \) is the absolute temperature, \( \nu \) is frequency, and \( \Delta \nu_T \) is the FWHM of the profile, given by

\[
\Delta \nu_T = \frac{\kappa}{2\pi} \sqrt{\frac{8kT \ln 2}{m}} \quad (8)
\]

In the above expression, \( k \) is the Boltzmann constant and \( m \) is the molecular mass.

Molecules and particles under these conditions, where the wavelength is small compared to the mean free path, are said to be in the Knudsen regime.

However, as the mean free path length decreases, the spectral distribution of the scattered light becomes increasingly governed by acoustically-driven disturbances present within the operating volume. This is referred to as the hydrodynamic regime.

These disturbances lead to the formation of two sharply-shaped peaks whose frequency shift from that of the incident source is reflective of the speed of sound in the medium [6,11]. Unfortunately, many conditions, including conditions at standard atmospheric temperature and pressure, correspond to an intermediate regime which must account for both random molecular motion and the correlated motion associated with acoustic propagation. Several models have been developed [9]; though the most widely used is the S6 model developed by Tenti et al. [10]. These models make use of a quantity defined as the \( y \)-parameter which is given as
\( y \equiv \frac{\lambda_s}{2\pi l_m} \approx \frac{NkT}{\sqrt{2\kappa v_o\eta}} \)  

where \( N \) is the number density, \( \lambda_s \) is the wavelength of the laser source, \( \eta \) is the shear viscosity, and \( v_o \) is a thermal velocity arising from the relation

\[ v_o = \frac{kT}{\sqrt{m}} \]  

It also is common for various values of \( y \) to be plotted as a function of a normalized frequency variable \( x \) defined as

\[ x = \frac{2\pi v}{\sqrt{2\kappa v_o}} \]  

to characterize a particular operating (Knudsen or hydrodynamic) regime. It is shown in Fig. 2 that the spectral shape of the Rayleigh scattering profile is highly determined by the \( y \)-parameter. It is evident that as the value of the \( y \)-parameter increases the lineshape transitions from the Gaussian distribution of the Knudsen regime to the grouping of three interconnected Lorentzian peaks associated with the hydrodynamic regime. It may be noted that for air at atmospheric pressure and a temperature of 298K, the value of the \( y \)-parameter is 0.798, indicating that the influence of acoustic contributions to the spectral profile cannot be omitted.
Figure 2. Rayleigh Scattering spectral lineshapes for a range of y-parameters.

It should be noted that while these models have proven reliable for simple diatomic gases (CO, N₂, etc.) they do not, as of yet, accurately predict the scattering profiles of more complicated molecules, such as the hydrocarbons that may be of interest to projects in turbulent mixing and combustion.

Spectral Filtering of the Rayleigh Scattering Signal

Spectrally filtering the Rayleigh-scattered light can serve a variety of purposes. With an accurate model of both the Rayleigh scattering lineshape and the profile of the molecular or atomic filter used, multiple parameters (such as temperature, pressure, and number density) can be extracted from the recovered signal which has passed through the
filter. Also, scattered light, which is in resonance with a particular molecular or atomic species may be absorbed greatly such that the transmitted signal (through the filter) is reduced by many orders of magnitude, thereby providing the potential for greatly reducing the signal associated with unwanted scattering from surfaces and/or large particles that typically interfere with and overwhelm gas-phase Rayleigh-Brillouin scattering (see Fig.1) It is the latter approach which will be the focus of the current work outlined within this thesis.
Chapter 2: Iodine Filter Cell Transmission Modeling and Validation

In order to obtain quantitative FRS measurements, the transmission profile of the molecular or atomic absorption filter, \( T(\nu) \), must be accurately known. Typically, this filter is comprised of a glass housing (or cell) filled with a specific atomic or molecular species, which is placed in front of a detector. Previously, in a variety of flow configurations, a molecular iodine filter has been used in conjunction with the 532nm (or 18788 cm\(^{-1}\)) output of a frequency-doubled Nd:YAG laser. This is a fairly common approach as there is a manifold of several molecular absorption features located in this frequency range as a result of transitions of the bound \( \chi(1\Sigma^+_g) \) and \( B(3\Pi_{0,v}) \) states [12].

The transmission profile, or conversely the absorption profile, of the cell varies as a function of cell pathlength (a generalized solution to Beer’s Law), as well as cell temperature and pressure via collisional and Doppler broadening [6].

The particular \( I_2 \) cell used in this work has a diameter of 76 mm and is 254 mm in length. It is wrapped in heating tape which is controlled by a temperature controller to within 0.1°C. The cell is operated under what has been termed a starved-cell arrangement [13]. In this configuration, the number density of iodine within the cell is set (typically at elevated temperatures) and the cell is sealed.

Alternatively, filter cells can be operated such that the number density can be varied by changing the temperature of a “cold-arm” built into the cell. This consists of a
small attachment to the cell body, typically cooled by a temperature controlled water
jacket. Iodine crystals are placed within the cold arm and by changing the temperature of
this portion of the cell, the crystals can be made to sublimate or evaporate, thereby
changing the number density of iodine within the main cell body [3,12]. By changing the
temperature of the cell, (or additionally the side arm temperature where applicable) the
transmission properties can be made to vary substantially. While this configuration
offers the maximum possible flexibility, the filter characteristics are a function of
temperature fluctuations within the side arm. For quantitative measurements using FRS,
the uncertainty in the filter transmission profile associated with even small side-
arm temperature fluctuations are unacceptable. Thus, in this work, the starved-cell
configuration was selected.

Transmission Modeling

In this work, the iodine absorption/transmission spectra is calculated using the
model developed at Princeton University [3,12]. The computational model, known
colloquially as the “Forkey code”, is included in Appendix A. Supporting documents can
be made available as necessary. This code allows for a calculation of cell transmission, or
$T(\nu)$, as a function of pathlength, temperature, and pressure.

Figure 3 shows the calculated effect of iodine vapor pressure on light transmission
for a cell with an absorption pathlength of 254 mm and cell temperature of 341 K. The
abscissa is presented in terms of Optical Depth, which is simply a logarithmic
representation of the transmission given by $OD = -\log_{10}(Transmission)$. 

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Figure 3. Iodine Cell Transmission as a function of iodine vapor pressure for a nominal cell temperature of 328K for a cell with pathlength of 254 mm
Figure 4. Iodine Cell Transmission as a function of cell temperature for a nominal cell pressure of 0.7 Torr for a cell with pathlength of 254 mm.

Figure 4 shows the effect of variation of cell temperature on light transmission. It should be evident from the figures above, that for a maximum optical depth, the iodine vapor pressure should be high, and the cell temperature as low as possible. However, in practice the optimal cell conditions will vary. Low cell temperatures typically will cause iodine crystals to form on the cell windows, which are the coldest areas of the cells and lead to optical obscurity. Also, high iodine vapor pressure within the cell, while substantially increasing OD, also leads to a broadening of absorption features which will
significantly reduce the amount of Rayleigh signal that is ultimately transmitted to the detector. For our particular cell, we found that the lowest temperature we were able to operate at consistently without iodine crystallization was 341K, and the cell pressure was set at 1.05 Torr, which served as a compromise between high optical density and sufficient transmission of the Rayleigh-scattered signal.

Experimental Verification using a Narrow Linewidth cw Laser

In order to experimentally verify the Forkey transmission code calculations and the iodine vapor pressure within our starved cell, scans across a pair of strong absorption features were performed utilizing a narrow linewidth (~5 kHz), continuous wavelength (cw), frequency-doubled Nd:YAG laser (JDS Uniphase NPRO M126N-1064-500). A schematic of the experimental configuration used is presented in Fig. 5.
Figure 5. Schematic of experimental configuration used for iodine spectral scans using a single-frequency, cw laser.

The output of the cw laser was approximately 500mW at 1064nm and was focused into a type-II KTP crystal for frequency doubling to generate 532-nm light. The 532-nm beam power was approximately 1mW following second-harmonic conversion. The beam was then reflected off of a series of mirrors with a 532-nm anti-reflective coating. In this manner, residual 1064-nm light, was separated and removed. The beam was then passed through an iris before being attenuated by a series of 60-40 beam-splitters and an ND filter. A reflection from the face of a beam-splitter was passed to a photodiode (PD, Thorlabs, DET10A, with a rise time of <1ns) in order to monitor the incident power. A
small amount of scattered light from the face of the ND filter (labeled as item 6 in figure 5) was collected in a single-mode optical fiber and transmitted to a high-resolution wavemeter (High Finesse WS-U). The wavemeter has an accuracy of 30 MHz and a spectral linewidth resolution of 75 MHz. A frequency-stabilized HeNe laser is coupled to the wavemeter and provides a calibration signal, which is updated every 10 minutes.

After passing through the final beamsplitter, the beam was focused through a series of removable, user-calibrated ND filters, a 532-nm notch filter, a temperature-stabilized I₂ cell, and onto a photomultiplier tube (PMT, Hamamatsu 1405CE). The output signals from both the photodiode and PMT were monitored on a digital oscilloscope (LeCroy Waverunner 104MXi) which has a bandwidth of 1GHz. Not shown in the schematic is a blanket of layered black cloth that was draped over both the I₂ cell and PMT, which served to mitigate any extraneous light and/or reflections that may otherwise impinge upon the PMT and lead to erroneous signal levels.

The frequency output of the cw Nd:YAG laser was thermally tunable over a specified range of 18786 cm⁻¹ to 18790 cm⁻¹. However, in practice, only a few regions of the tuning range were able to maintain stable, mode-hop-free operation for long periods of time. It should be noted that a sudden shift in frequency, when initially tuned to a frequency corresponding to an iodine absorption line, can lead to an equally sudden increase in signal level at the PMT (or in the case of later studies, a CCD chip). This increase may be several orders of magnitude and, therefore, potentially damaging. It is absolutely necessary to ensure stable frequency operation of the laser system when performing FRS measurements. In the present case, stable operation was achievable over
a tuning range from 18786.6 to 18788.1 cm\(^{-1}\), allowing access to the strong R86(33,0)/R44(39,2) and R106(34,0) transitions near 18787.3 cm\(^{-1}\).

In performing a scan across these absorption features, the frequency of the Nd:YAG laser was slowly adjusted and recorded by the wavemeter, which has a relative accuracy of ±5 MHz. Signals at the photodiode and photo-multiplier tube were integrated over 500 ms on the oscilloscope, reducing statistical noise to negligible levels. As the frequency of the laser varied over the absorption feature, ND filters (item 9 in figure 4) were added or removed as necessary to maintain signals in the linear range of the PMT. Removing an ND filter of known OD is equivalent to rescaling the measured transmission by 10\(^{\text{OD}}\). The results of this study are presented in Fig. 6.
Figure 6. Measured (closed symbols) transition profile over the R86(33,0)/R44(39,2) and R106(34,0) transitions near 18787.3cm⁻¹ are shown along with the modeled transmission profile (solid line) generated utilizing the model developed by Forkey et al. [3,12]. Cell conditions correspond to 353K, 1.05 Torr, and a 254mm pathlength.

To account for the continuous I₂ background absorption (peak transmission when not tuned to an absorption feature is not identically unity) a normalization was performed in which the measured signal was matched to the theoretical model value at a non-resonant frequency, which for the current work was ~18787.4cm⁻¹. Signal-to-background
levels were recorded as 20 to 185 depending on the overlap between the laser frequency output and the spectral profile of the Iodine absorption feature. Peak optical density measurements were measured at 7.0, compared to the OD of 7.19 predicted by the model for the R86(33,0)/R44(39,2) transition, and a match of 4.3 for the R106(34,0) transition. These results are similar to the findings of Forkey et al. [3,12] which report an accuracy of ±10% for absorption features in this spectral range.

The results are important for two reasons. First, this measurement validates the experimental methodology as results with sufficiently high signal-to-background were recorded over seven orders of magnitude. Secondly, the theoretical modeling of the iodine absorption spectra can be modeled effectively for a cell operating under our experimental conditions.
Chapter 3: Brief Review of Previous FRS Methodologies and Applications

The filtered Rayleigh scattering diagnostic has proven to be a valuable tool in a variety of applications, often enabling the acquisition of quantitative information typically obscured by additional intensely-scattering processes. One application of FRS has been qualitative and quantitative velocity measurements [e.g., 14-28]. As noted in chapter 1, the FRS signal undergoes a Doppler shift proportional to the velocity of the scattering media. For example, in work done by Miles et al., [14,15] CO₂ condensates, formed in a Mach 3 flow passing over a wedge, exhibit small-particle scattering in the Rayleigh regime that is Doppler shifted corresponding to their velocity. By tuning the interrogating laser such that its spectral frequency coincides with a molecular absorption feature (of I₂ in this case), background scattering from the windows and walls of the wind tunnel, which might otherwise dominate the collected signal, can be suppressed. However, the Doppler-shifted scattering from the condensates in the flow will, under the correct filter conditions, fall spectrally outside the molecular absorption feature and can be transmitted to a detector. By varying the frequency of the interrogation laser within the bandwidth of the I₂ absorption feature, high or low velocity regions of the flowfield can be highlighted [14,15]. This technique often is referred to as Planar Doppler Velocimetry (PDV) [16] and has been utilized in flow configurations including compressible free
shear layers [17], compressible mixing layers, rotator wakes, sonic and supersonic jets, low speed flows and others [e.g., 18-28].

The same Doppler shifting holds true for molecular scattering (as opposed to particle scattering) as well, but additional broadening of the scattered profile associated with random molecular motion can provide information on additional flow parameters. Typically, the Doppler broadening of the Rayleigh profile is used to determine temperature in particle-laden flows. For example, Hoffman and Leipertz [29] demonstrated the utility of FRS for 2D temperature measurements in a sooting premixed, methane/air flame. Subsequently, several research groups used FRS and particle imaging velocimetry (PIV) diagnostics for simultaneous temperature and velocity field measurements in combustion environments [e.g., 13, 30-32]. Elliot et al. [13] applied simultaneous FRS/PIV to stagnation flames and Most and Liepertz [30] utilized FRS/PIV diagnostics to investigate lean methane-air V-flames. Similarly, Most and coworkers [32] applied joint FRS and PIV imaging techniques to turbulent flames, thereby enabling qualitative studies of flame structure interactions with flow vortices and strain rate and enabled a quantitative determination of the Farve mean turbulent flux ($u^\prime Y$), an important parameter in turbulent combustion modeling [33]. Here, $Y'' = Y_1 - Y$ is a measure of the fluctuation, $\overline{\rho Y}/\overline{\rho}$ is the Favre mean average of scalar $Y$, $u$ denotes the velocity, and $\rho$ is the gas density. Kearney et al. [31] obtained temperature measurements in non-premixed flames through a joint FRS and spontaneous Raman scattering diagnostic. In this work, the species information obtained via spontaneous Raman scattering enabled a more accurate determination of the local composition which
is needed to convert FRS signal to temperature due to the dependence of the signal on the local Rayleigh scattering cross section and spectral lineshape (see Eq. 1). This approach, when combined with a flamelet-based model correlating the FRS and Raman signals, allowed for temperature determination with a stated accuracy of ±6% [31]. The same researchers also have applied FRS in a number of additional configurations including heated air jets, premixed flat flames, vortex driven diffusion flames, and additional sooting flames [32].

In addition to temperature, measurements of pressure and density can be made utilizing the FRS technique [e.g., 3]. As an example, Boguzko and Elliot [6, 7] measured average temperature, pressure and density using FRS in compressible unseeded flows and thermal fields by use of a frequency scanning technique [6, 7]. A schematic taken from their work is presented below in Fig. 7.
Figure 7. Demonstration of the FRS scanning technique. (a). A schematic of the laser and Rayleigh scattering profiles convoluted with the absorption spectra of iodine as the laser frequency is scanned across a strong absorption feature. (b). FRS signal as a function of laser frequency for several experimental temperatures. Figure and caption taken from [7].

Figure 7 clearly demonstrates the relationship between the spectral profile of the scattering signal and the thermodynamic properties of the flowfield. This methodology has been applied to various statically-steady flows including a transonic jet (Mach numbers of 0.8-1.5) and laser-induced energy deposition in quiescent air [e.g., 6,7].
However, in all the aforementioned studies, pulsed, injection-seeded, Nd:YAG lasers were used as the illumination source and a common limitation was encountered – the measured suppression of unwanted scattering signals was much less than predicted by theory. Specifically, extinction ratios were several orders of magnitude lower than predicted by the model developed by Forkey [3]. It should be noted that the model developed by Forkey was verified by measurements using single-frequency, cw lasers (similar to the results shown in Fig. 5 of this thesis).
Chapter 4: Spectral Purity Effects on FRS Measurements

In Chapter 2 it was shown that for true single-frequency cw laser sources with very narrow spectral linewidths (~kHz), the theoretical I$_2$ absorption model matches measured values, which is consistent with the previous experimental work of Forkey [3]. However, for pulsed, frequency-doubled, injection-seeded, Q-switched Nd:YAG lasers, the maximum reported level of attenuation has been limited to $10^3$ to $10^4$ regardless of the theoretical optical depth for a given set of cell conditions [6,12,33,34]. This relatively high level of transmission can limit the application of the FRS technique due to the limited suppression of potentially unwanted Mie or surface scattering. In fact, this has been one of the limiting factors of why FRS has not realized the enormous potential evident upon its introduction almost twenty years ago. To obtain quantitative measurements using FRS, one has to be sure that the signal at the detector is the result of Rayleigh-Brillouin scattering alone; therefore, it is necessary that the mechanism limiting unwanted scattering suppression be thoroughly understood and if possible, mitigated.

It is generally accepted that the suppression limitations which arise when performing FRS with pulsed, injection-seeded, laser sources are a result of non-ideal spectral purity of the laser output. Specifically, in an injection-seeded Nd:YAG laser, a very narrow linewidth (~10kHz) cw laser is used to preferentially populate one desired longitudinal mode within the laser cavity, thereby “seeding” that mode. As the laser pulse
forms, this pre-seeded mode dominates competing, cavity induced modes which initiate from noise within the cavity. The resulting output is a laser pulse which is predominantly comprised of a single longitudinal mode, and therefore has a very narrow spectral bandwidth or laser linewidth. To facilitate a good match between cavity modes and the seed laser, the laser cavity is “dithered” or changed slightly in length, typically by means of a piezo-controlled resonator mirror. The modes, whose spacing (Δν) is determined by the length of the cavity through the relation Δν = c/2L where L is the cavity length and c is the speed of light, can then be shifted spectrally to nearly match the frequency of the seed laser. In injection-seeded lasers, the seed laser does not have to exactly match the resonance of the slave laser, only a portion of the optical power needs to overlap with one resonator mode, thus there may be some mismatch between the seed laser frequency and the pulsed laser output. With this process, the pulsed laser output effectively “locks”\(^2\) to a single mode with an efficiency greater than 99% leading to a nearly Fourier transform limited output bandwidth of <0.003cm\(^{-1}\) near 1064nm.

However, a small fraction of energy is contained in other supported cavity modes due to residual mode competition. Thus, the spectral laser output can be approximated as a narrow spectral peak comprising the vast majority of the energy, superimposed on a low-energy quasi-broadband pedestal. While this may be of little consequence to many traditional diagnostics, the implications to FRS are substantial as the broadband pedestal is much spectrally broader, than the I\(_2\) absorption features used to reject unwanted Mie or surface scattering. Therefore, the scattering from photons of these frequencies fall outside

\(^2\) The term “lock” is used colloquially and is not meant to be confused with injection locking, where the seed laser frequency exactly matches a particular longitudinal mode and the frequency of the pulsed output.
of the I₂ absorption band and pass through the cell with little attenuation. Despite containing only a small portion of the total energy, scattering from surfaces or particles may be many orders of magnitude greater than Rayleigh scattering and can easily lead to signal levels comparable to or greater than that of Rayleigh scattering reaching the detector, thus reducing the effectiveness of the I₂ cell and limiting FRS application. In this manner, the effective attenuation realizable with a combination of an I₂ filter cell and frequency-doubled, injection-seeded Nd:YAG laser is limited by the amount of energy contained in this broadband, non-seeded spectral component.

Laser Linewidth Effects

Before considering the specific effects of broadband pedestal, we will first examine the role of laser linewidth, since for a pulsed laser, the spectral bandwidth is finite, which is limited by the “time-bandwidth product” of the pulse. Linewidth effects must be understood first in order to decouple them from spectral impurity effects. To put this in perspective, consider the case where the spectral bandwidth is sufficiently large such that portions of the spectral profile fall outside of the I₂ absorption feature (typically on the order of 1 GHz). For this case, the linewidth of the laser will limit the effectiveness of the filter in much the same way as a broadband pedestal. Figure 8 shows a calculation of the effect of increasing laser linewidth on realizable attenuation through an iodine cell (top). The effective optical density is calculated as the convolution of an assumed laser spectral distribution and the theoretical iodine spectral model from Forkey et al. [3]. The spectral distribution of the laser is modeled as a Gaussian distribution with
a full width at half maximum (FWHM) defined as the laser linewidth ranging from 75 to 400 MHz. For these calculations, the laser spectral distribution is convolved with the R56(32,0) and P159(39,0)/P103(34,0)/P53(32,0) transitions of the B(3^3Π_{0+u})←X(^1Σ^+_g) electronic system of I₂. In these calculations, the model inputs have been chosen to match our experimental cell conditions, namely, an optical pathlength of 254mm, a cell temperature of 341K and cell pressure of 1.05 Torr. The bottom portion of Fig. 8 shows the maximum attenuation (the minimum transmission) at 18788.33 and 18788.44 cm⁻¹, corresponding to the spectral “peaks” of the absorption features. It is clear that the realizable attenuation, as determined as the optical density of a given I₂ absorption peak, is not significantly affected for laser linewidths less than 200 MHz.
Figure 8. (Top) Calculations of the effective transmission profile of the iodine cell as a function of laser linewidth. Cell conditions correspond to experimental conditions: cell temperature of 341K, cell pressure of 1.05 Torr, cell length of 254mm. The spectral scans are a result of the convolution of the laser spectral profile with the iodine absorption profile. (Bottom) Maximum optical density as a function of laser linewidth.
Spectral Purity Effects on Iodine Cell Transmission

As shown above, the effect of finite laser linewidth has little effect on iodine cell attenuation for linewidths less than 200 MHz. The injection-seeded laser utilized in our measurements (Spectra Physics PRO 290) has a pulse duration of ~ 6 ns at 532 nm, and subsequently a Fourier-limited laser spectral linewidth of <75 MHz (based on a $\sqrt{2}$ scaling of the fundamental output characteristics). This value also is consistent with linewidth measurements using a high-resolution wavemeter. Therefore, any decrease in the effectiveness of the iodine cell in terms of resonant scattering attenuation can be attributed to energy contained within the broadband pedestal. This spectral distribution of energy within a laser pulse can be defined in terms of spectral purity; that is, the ratio of the energy contained within a single longitudinal mode to the total energy output. In this section, calculations demonstrating the effects of decreasing laser spectral purity (or equivalently an increasing amount of energy contained in the broadband pedestal) on realizable $I_2$ cell attenuation are presented.

The seeded portion of the laser spectral output, $L_S(\nu)$, is modeled as a Gaussian distribution with a FWHM of 75MHz. For cases where the spectral purity is less than 1, the broadband pedestal, $L_B(\nu)$, is modeled as a Lorentzian distribution with a FWHM of 42GHz [12, 35] which would be expected from the frequency-doubling of the fundamental output at 1064 nm with a FWHM of 30 GHz. A 30-GHz bandwidth typically is reported for the majority of Nd:YAG lasers from major manufacturers and is the reported FWHM for the laser used in the current experiments. This modeled bandwidth also is consistent with the 40 GHz (FWHM) bandwidth of residual
(broadband) light from a Nd:YAG laser measured by Seasholtz and Buggele [34] using an I₂ cell and Fabry-Perot interferometer. The spectral distribution describing the total laser output is represented as

\[ L_T(\nu) = L_S(\nu) + \alpha L_B(\nu) \]  \hspace{1cm} (12)

where \( \alpha \) is a free parameter which may be varied to simulate varying fractions of energy contained within the broadband pedestal of the laser spectral profile. For the cases of spectral purity \(< 1\), the magnitude (\( \alpha \)) of the Lorentzian distribution describing the broadband, non-seeded component is successively increased such that the spectral purity decreases. More specifically, increasing the portion of the non-seeded Lorentzian distribution, \( L_B(\nu) \), effectively models an increasing fraction of energy being dispersed to additional longitudinal modes near the desired “seeded” longitudinal mode.

Consistent with previous definitions [36,37], the spectral purity, \( SP \), of the laser output is defined as

\[ SP = 1 - \frac{\int_{\nu_1}^{\nu_2} L_B(\nu) \, d\nu}{\int_{\nu_1}^{\nu_2} L_S(\nu) \, d\nu} \]  \hspace{1cm} (13)

where \( \nu_1 \) and \( \nu_2 \) are lower and upper frequency limits of integration, respectively, sufficient to capture the full Lorentzian profile of the broadband portion of the laser gain profile. Figure 9 shows example distributions of \( L_T(\nu) \) for varying values of \( \alpha \). It should be noted that while the broadband portion of the laser spectra is modeled as a continuous Lorentzian distribution, it is in fact not continuous. As mentioned previously, the laser cavity supports distinct longitudinal modes, spaced at frequencies corresponding to \( c/2L \), where \( c \) is the speed of light and \( L \) is the cavity length. For the laser in use with the
current measurements, the spacing of these discrete modes is \(~200\ \text{MHz}\). A small packet of individual longitudinal modes is shown in Fig. 9. The spacing of the individual modes is very small compared to the total bandwidth of the broadband component. In addition, the apparent “structureless” features of the iodine absorption spectra outside of the strong R56(32,0) and P159(39,0)/P103(34,0)/P53(32,0) transitions of interest, allows for these discrete modes to be modeled continuously without a significant loss of validity in the calculation results.

Figure 9. Example laser spectral distributions, $L_T(v)$, for a variety of spectral purities, SP. Individual longitudinal modes, which comprise the broadband pedestal, are shown as well for a spacing of 200MHz. In subsequent calculations, the ensemble of discrete modes is modeled as a continuous Lorentzian distribution.
Results showing the convolution of the iodine absorption spectra, $T(\nu)$, with the modeled laser spectral distribution, $L_T(\nu)$, are shown in Fig. 10. Prior to the convolution operation, $L_T(\nu)$ is properly normalized such that $\int_{\nu_1}^{\nu_2} L_T(\nu) = 1$. For these calculations, cell conditions are modeled to match the experimental conditions; that is, a cell temperature of 341K, a cell pressure of 1.05 Torr, and a cell pathlength of 254mm. Example spectra (shown in terms of optical density) are shown for SP values of 1.0 (no broadband component), as well as 0.999999, and 0.999.

![Spectral Scans](image)

Figure 10. Calculations showing the effects of decreasing spectral purity (SP) on I$_2$ cell transmission for a laser linewidth equal to 75 MHz. Conditions correspond to a cell temperature of 341K, cell length of 254 mm, and cell pressure of 1.05 Torr. The spectral scans represent the convolution of the laser spectral distribution, $L_T(\nu)$ and the I$_2$ absorption spectra over the R56(32,0) and P159(39,0)/P103(34,0)/P53(32,0) transitions of the B←X electronic system of I$_2$. 

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These results are striking, as a change in SP from 1 to 0.999999 results in a change in the maximum attainable optical density from 9.6 to 6.1, a change of greater than three orders of magnitude when only 0.0001% of the laser energy is contained in the broadband pedestal. A further reduction in SP to 0.999 leads to a maximum attenuation of only 3.1, comparable to the results of previous studies utilizing pulsed, injection seeded, Nd:YAG laser systems [3, 33, 34]. Furthermore, calculations show that to approach the theoretical attenuation associated with a spectrally pure source (SP=1), a SP of greater than 0.999999999 (or one part per billion in the broadband pedestal) is required for the current cell conditions and for a seeded bandwidth of 75 MHz.

Experimental Measurement of Iodine Cell Attenuation

A schematic of the experimental setup used to measure the transmission profile of the current iodine cell is presented in Fig. 11. This is similar to the setup used for the cw laser scans in chapter 2. Briefly, the frequency-doubled output of an injection-seeded, Q-Switched, Nd:YAG laser is passed through a combination of a λ/2 waveplate and thin film polarizer. By adjusting the orientation of the waveplate, the laser power can be attenuated as necessary to maintain operation in the linear regime of the detection devices (photodiode and photomultiplier tube). The beam then passes through an iris to minimize the effects of spatial variation in optical frequency inherent to injection-seeded, Q-Switched Nd:YAG laser systems [38]. Backscatter from a 532nm mirror is collected using a single mode optical fiber and directed to the High Finesse WS-U wavemeter for
frequency monitoring and recording. The beam then passes through another iris, and a series of beamsplitters and ND filters to further attenuate the laser energy. Individual laser pulse power is monitored using a photodiode (PD, Thorlabs DET10A, rise time <1 ns). The beam is then focused through a set of user-calibrated ND filters described in chapter 2, a 532nm notch filter, the I₂ cell, and onto a photomultiplier tube. The output of the photodiode and photomultiplier tube is monitored on a digital oscilloscope (LeCroy Waverunner 104MXi).
Figure 11. Schematic of experimental setup used to perform I₂ scans
The output of the Q-switched, pulsed Nd:YAG laser can be frequency tuned by adjusting the temperature of the internal Nd:YVO₄ seed laser. The minimum frequency step using this laser source was ~2 MHz based on the minimum possible temperature adjustment of 0.001K. However, in practice, the laser was frequency tuned in steps of ~50 MHz across the iodine absorption spectrum near 18788.3 cm⁻¹, a region corresponding to the strong R56(32,0) and P159(39,0)/P103(34,0)/P53(32,0) transitions of the B←X electronic system of I₂. At each frequency position, 250 instantaneous transmission measurements were taken. Functionally, this measurement is expressed as

\[ T = \left( \frac{P_{MT} - P_{MTBG}}{P_{PD} - P_{PD_{BG}}} \right) \cdot T_{NR}^{-1} \cdot N D_S \]  

(14)

where \( T \) is the transmission through the I₂ cell, PMT, and PD the voltage readings of the respective devices, and PMT_{BG} and PD_{BG} the background signals acquired with the laser path blocked. \( T_{NR} \) is the ratio of the PMT and PD signals acquired at a non-resonant frequency (the scaling factor of chapter 2 was used to account for any DC offset resulting from losses or broadband absorption or alternatively the transmission ratio at non-resonance) and ND_S is a scaling factor that accounts for the addition or subtraction of any combination of the user-calibrated ND filters. Typical standard deviations were ~3% of the mean value.

Experimentally, the linewidth of the laser was determined to be < 75MHz, based on the minimum resolution of the wavemeter, and also consistent with a Fourier-transform-limited pulse of ~6 ns which is the measured pulse duration of the 532-nm output. In measuring the laser linewidth, the spectral linewidth of a series of \( 10^4 \) individual laser pulses was recorded on the wavemeter. Over this interval, 93% of the...
pulses were recorded as having a linewidth of <75MHz (the minimum resolution of the device), with the remaining 7% of the readings falling between 75 and 100 MHz. Note that, consistent with Fig. 8, the measured I\textsubscript{2} cell attenuation should be largely unaffected by a variation in linewidth between 75 and 100 MHz.

Figure 12 shows results of one scan across the R56(32,0) transition near 18788.33 cm\textsuperscript{-1}, taken with cell conditions corresponding to a temperature of 341K, a cell pressure of 1.05 Torr, and a cell length of 254mm. Also shown is the theoretical model of the I\textsubscript{2} absorption spectrum in this region [12]. Consistent with previous studies, our measured transmission values are several orders of magnitude higher than the model suggests [3, 33, 34]. Using the results presented earlier, we can calculate an estimate of our laser spectral purity based on the finite absorption measured through our cell. For a laser with a nominal linewidth of 75 MHz (FWHM), an OD of ~5.1 for these cell conditions, would imply a spectral purity of ~ 0.999991.
Figure 12. Measured transmission profile over the R56(32,0) transition near 18788.3 cm\(^{-1}\) using an injection-seeded, Q-switched, pulsed laser. Also shown is the theoretical absorption profile for a cell operating at our conditions.

Seed Laser Power Effects on Spectral Purity

As shown above, for many laser diagnostic systems, the spectral purity of the laser output is a fundamental limiting factor to the applicability of filtered Rayleigh scattering. As a means of assessing one potential methodology for increasing the spectral purity of our laser system, we investigated the effect of seed laser power on spectral purity. As described earlier, the spectral purity of a laser source is defined as the ratio of
the energy contained within a single longitudinal mode to that of the total pulse energy.

Detailed theoretical descriptions of injection-seeding have been presented within the literature [e.g., 35, 36] and as described by Barnes and Barnes [36], spectral purity (SP) is a strong function of the circulating seed pulse energy ($E_{seed}$):

$$E_{seed} = \frac{E_N}{T_R C_o^2} \frac{SP}{1-SP}$$  \hspace{1cm} (15)

In the previous equation, $E_N$ is the “noise,” or energy contained in the unseeded cavity modes and $T_R C_o^2$ is a measure of the coupling between the seed laser output and the laser oscillator. Typically, seed lasers operate continuously; therefore $E_{seed}$ can be formulated as $P_{seed} \Delta t$, where $P_{seed}$ is power of the seed laser beam within the laser cavity, and $\Delta t$ the pulse duration governed by the laser cavity round trip time. As noted from the previous equation, as the desired SP approaches unity, the seed laser power increases to infinity.

To study these effects within our laser system, a series of user-calibrated ND filters were inserted after the output of the seed laser, thus reducing the power which was coupled into the laser oscillator cavity. Special care was taken to ensure that the alignment of the optical path was not affected by the insertion of the filters. Scans across the R56(32,0) transition of $I_2$ were then repeated for a variety of adjusted $P_{seed}$ values, ranging from 1mW to 15mW. In practice, seed laser powers of less than 1mW resulted in unstable seeding, multi-mode operation, and mode-hopping (potentially damaging effects). An additional concern was that of frequency drift over the course of the measurement. When preforming a scan across the $I_2$ spectra, 200 shot averages were taken at each spectral location. If the instantaneous output frequency of the laser deviates
from the mean frequency by a significant amount, an appreciable difference in the transmission/absorption value may result, thus corrupting the interpretation of the seed-power effects. To investigate this possibility, long, 10,000 pulse recordings were performed for seed laser powers of 1.5, 7.5, and 15 mW. Histograms of the measured frequency values as, a deviation from the mean frequency, are presented in Fig. 13.

![Histograms of the output frequency fluctuations (with respect to the mean output frequency) for circulating seed powers of 1.5, 7.5, and 15 mW. Also shown is the measured frequency fluctuation of the cw 1064-nm seed laser, which is near the resolution of the wavemeter.](image)

The histograms for the 1.5 and 7.5 mW are very similar, with a FWHM of approximately 50 MHz. The 15 mW seed power case was slightly broader with a FWHM of <100 MHz.
This can be thought of as an artificial broadening of the linewidth of the laser pulse to <125 MHz for all cases; however, as was shown previously (see Fig. 8), a linewidth change of this magnitude should have little effect on the achievable optical density.

Shown in Fig. 14 is the measured peak optical density of the R56(32,0) transition of I₂ for the two cell temperatures of 341 K and 353 K as a function of circulating seed power, which varies from ~ 1 mW to 15 mW. The optical densities recorded range from ~ 4.0 at a seed laser power of ~1 mW to 5.3 for a seed laser power of ~15 mW. Over this span, there is a logarithmic relationship between the coupled power of the seed laser and the maximum attainable optical density. Therefore, for an order of magnitude increase in seed laser power, there is a corresponding order of magnitude increase in optical depth.
Figure 14. Measured peak optical density of the R56(32,0) transition near 18788.3 cm$^{-1}$ using the injection-seeded, Q-switched, pulsed laser for circulating seed powers ranging from 1 mW to 15 mW. Measurements are performed for two cell temperatures corresponding to $T_c = 341$ K and $T_c = 353$ K. The “dotted” line through the data represents a “best fit” through both sets of data and is intended in aiding visualization only.
Figure 15. Measurements of spectral purity as a function of circulating seed power (1 to 15 mW) using the optical density measurements reported in Fig. 14 and the relationship between optical density and spectral purity. Also shown is the theoretical relationship between circulating seed power and spectral purity as given by Barnes and Barnes [36] and Eq. (3). Note the excellent agreement between the circulating seed power and the estimated spectral purity determined from the measured optical densities.

Fig. 15 reports the estimated laser spectral purity as a function of circulating seed power, as determined from the measured optical densities (Fig. 14) and the relationship between optical density and spectral purity defined from Fig. 10. As shown in Fig. 15 there is a linear relationship between the logarithmic value of (1-SP) and the logarithm of laser seed power. Also shown in Fig. 15 is a curve depicting the theoretical scaling of Barnes
and Barnes [36] as described in Eq. (15). In order to construct the curve, the value of $E_N/T_R C_o^2$ was determined at one seed power ($P_s = 15$ mW) and one cell condition ($T_c = 341$K) and used for remaining seed power measurements at both cell temperatures. It is evident from the results of Fig. 15 that for the conditions in this study, the theoretical relationship between circulating seed power and laser spectral purity holds. These results imply that an increase in seed laser power directly results in an increase in laser spectral purity. Therefore, one potential methodology for increasing laser spectral purity, and subsequently increasing the suppression capabilities of the FRS technique, is utilizing higher powered seed lasers. However, it should be noted that the effects of coupling high powered seed lasers into the operating cavity of a laser system have not been studied extensively.
Chapter 5: Implementation of an external Fabry-Perot Etalon as a Means of Increasing Spectral Purity

As demonstrated in the previous chapter, a limiting factor of the FRS diagnostic technique is laser spectral purity. Commercially-available, injection-seeded, Q-switched laser systems exhibit a non-idealized output as it pertains to spectral purity. This manifests itself as laser output which is described effectively as a spectrally-narrow peak superimposed on a spectrally-broadband, low-energy pedestal.

This chapter will demonstrate one potential method to increase the spectral purity of the laser beam is to reduce the energy contained within the pedestal by spectrally-filtering the laser output with an external Fabry-Perot interferometer (FPI) or etalon. Specifically, a FPI or an etalon consists of two parallel highly reflecting surfaces used to form a multiple-beam interference device. An etalon is one substrate which consists of two highly reflective surfaces, while an interferometer consists to two parallel mirrors. For the remainder of this thesis, the term “etalon” will be used to describe both a “Fabry-Perot interferometer” and a “Fabry-Perot etalon”. When constructive interference occurs within the etalon, transmission is maximized. This interference is a function of the optical pathlength between reflecting surfaces; therefore, by slightly changing the effective distance between the reflecting surfaces of the etalon, a bandwidth of target frequencies can be preferentially selected to transmit through the etalon, while other wavelengths are
suppressed. For FPIs, the distance between the two mirrors can be adjusted through piezo-electric actuation or for both FPIs or true etalons, the effective distance between the reflecting surfaces can be changed by slightly adjusting the angle between the “etalon” and incident laser beam. The latter approach was taken in the current study. By careful selection of cavity spacing and reflectivity, the etalon can be made to serve as a highly sensitive, ultra-narrowband bandpass filter. This will be shown explicitly in the upcoming section.

Previously, intra-cavity etalons, (an etalon placed within the laser cavity) have been suggested as a method to diminish energy contained in the broadband pedestal and therefore increase suppression of unwanted scattering (as there will be less available energy in frequency bands that fall outside the molecular absorption band of the FRS cell) [34]. Using this methodology, reported suppression capabilities, or maximal optical depth, increased by approximately one order of magnitude. However, due to the fluctuating (thermal) conditions occurring within a laser cavity, the intra-cavity etalon is extremely difficult to use, requiring sophisticated temperature stabilization and frequency tuning. Our approach attempts to spectrally filter the frequency doubled 532-nm output externally (as opposed to the fundamental 1064-nm oscillator beam internally) as the beam propagates to the experimental measurement volume.

Etalon Transmission Formulae

As noted in the preceding section, the degree to which incident light is transmitted through a FPI or etalon is a function of cavity spacing, surface reflectivity, and optical
pathlength (experimentally realized as tuning angle). As shown in Fig. 16, an incident ray of light undergoes multiple reflections within the etalon cavity. At each interface, a portion of the electric field is transmitted and reflected.

The phase difference associated with each double-pass of the cavity is given by

$$\varphi = 4\pi \mu d \cos \theta / \lambda_o$$  \hspace{1cm} (16)
where $\mu$ is the index of refraction, $d$ is the cavity spacing, $\theta$ the tuning angle, and $\lambda_o$ the wavelength of the incident light. The amplitudes of the transmitted waves, following the notation presented in Hecht [39], is given by

$$E_{1t} = E_oo't't'e^{i\omega t}$$  \hspace{1cm} (17)

$$E_{2t} = E_oo't't'\rho^2 e^{i(\omega t-\varphi)}$$  \hspace{1cm} (18)

$$E_{3t} = E_oo't't'\rho^4 e^{i(\omega t-2\varphi)}$$  \hspace{1cm} (19)

$$E_{Nt} = E_oo't't'\rho^{2(N-1)} e^{i[\omega t-(N-1)\varphi]}$$  \hspace{1cm} (20)

A summation of these amplitudes yields the equation

$$E_t = E_o e^{i\omega t} \left[ \frac{tt'}{1-r^2e^{-i\varphi}} \right]$$  \hspace{1cm} (21)

In the above equations, $E_o t$ denotes the amplitude of the transmitted field, and $E_o r$ the reflected field. $E_o e^{i\omega t}$ is the equation of the incident wave. Noting that for self-similar surface coatings, transmission and reflectivity are equal for both faces; that is $t=t'$, $r=r'$. In this manner, the transmitted intensity is obtained by multiplying $E_t$ (the total transmitted electric field) by its complex conjugate. Denoting the transmission and reflection of the etalon surfaces as $tt'=T$, and $r^2=R$, respectively, the intensity of the transmitted light is derived as follows:

$$I_t = T^2/[(1+R)^2 - 2R \cos \varphi]$$  \hspace{1cm} (22)

$$I_t = T^2/[(1-R)^2 + 4R \sin^2(\varphi/2)]$$  \hspace{1cm} (23)

$$I_t = \frac{R^2}{(1-R)^2} \left( \frac{1}{1+[4R/(1-R)^2] \sin^2(\varphi/2)} \right)$$  \hspace{1cm} (24)

$$I_t = [T/(1-R)^2] \cdot [1 + F\sin^2(\varphi/2)]^{-1}$$  \hspace{1cm} (25)

$$I_t = [T/(1-R)^2] \alpha(\varphi)$$  \hspace{1cm} (26)
where \( F = 4R/(1 - R)^2 \) is defined to be the finesse of the system and

\[
\mathcal{A}(\varphi) = [1 + F \sin^2(\varphi/2)]^{-1}
\]  

(27)

is known as the Airy function [39-42]. When \( \varphi/2 \) is a multiple of \( \pi \); that is, when \( \varphi = 2\pi n \) (where \( n \) is an integer) the Airy function is equal to unity, and the transmitted intensity is at a maxima. As \( \varphi \) varies from \( 2\pi n \), the transmitted intensity decreases by an amount governed by the finesse, \( F \).

A plot of the transmission as a function of \( \varphi \) is presented in Fig. 17 for various values of finesse. Note that for very specific values of \( \varphi \), the transmitted intensity is equal to the incident intensity, regardless of the reflectivity of the surfaces of the etalon cavity.
Figure 17. FPI/etalon transmission is maximized for values of $\phi = 2\pi n$. The value of the transmission minima, occurring at $\phi = \pi n$, is a non-linear function of the Finesse, $F$.

However, in practice we adjust or ‘tune’ the etalon transmission by varying the angle ($\theta$) at which the incident light strikes the FPI or etalon. Figure 18 shows the transmitted intensity for an etalon with a finesse of 250, and with a fixed cavity spacing of 5 mm as $\theta$ is varied. It is clear from the figure that the transmission is highly sensitive to tuning angle, and in practice, achieving high values of transmission may be non-trivial.
In the current work, we are using a 20-mm clear-diameter, air-spaced etalon with a 5-mm thick, Schott Zerodour (Grade 0) spacer such that ambient temperature fluctuations result in negligible bandpass fluctuations. The surface reflectivity is at 88\textpm{}2\% and the bandwidth is 1.4 GHz. Due to limitations in the manufacturing process, the cavity parallelism is limited to $\lambda/100$ and the maximum transmission is approximately 95\%. A plot of the transmission as a function of wavenumber is shown in Fig. 19 using the specifications of our specific etalon, and for a tuning angle set to correspond to allow for maximum transmission in the region of a strong iodine absorption feature.
Figure 19. Transmission of the etalon as a function of wavenumber.

Light in the frequency (or equivalently wavelength) range outside the bandpass of the etalon (presumably the broadband pedestal of the pulsed laser output) will experience a transmission of < 0.5%, or an equivalent OD of 2.3. In this manner, by carefully tuning the etalon with respect to the incident laser light, a significant fraction of the energy contained in the broadband pedestal of the beam may be effectively diminished by several orders of magnitude. By convolving the etalon transmission profile with the
model of our laser output, $L_T(\nu)$, we effectively model a new lineshape, $L_e(\nu)$. In comparing the spectral power distribution of both of these profiles, we can estimate the effective change in spectral purity as a result of implementation of the etalon. Performing this calculation yields an estimated spectral purity of 0.99999946 for the beam which has been modified by the etalon, as compared to a spectral purity of 0.999991. Figure 20 clearly demonstrates the reduction in energy contained in the broadband portion of the laser spectra.
Figure 20. The laser output, $L_T(\nu)$, is shown against the profile transmitted through the etalon, $L_{te}(\nu)$. The tuning angle has been set such that transmission is maximized at a wavenumber corresponding to the R56(32,0) transition near 18788.3 cm$^{-1}$.

In convolving the $L_{te}(\nu)$ profile with a modeled iodine absorption spectra with cell conditions set to match those used in the experiment, (cell temperature and pressure of 341K and 1.05Torr, and pathlength of 254mm) it is seen that the predicted transmission is effectively decreased by more than one and one-half orders of magnitude from an OD of 5.2 to 6.5. This is shown in figure 21.
Figure 21. Comparison of attenuation with and without use of the external etalon.

Conditions correspond to a cell temperature of 341K, cell length of 254 mm, and cell pressure of 1.05 Torr. The spectral scans represent the convolution of the laser spectral distributions, $L_t(v)$, and $L_e(v)$ and the I$_2$ absorption spectra over the R56(32,0) and P159(39,0)/P103(34,0)/P53(32,0) transitions of the B←X electronic system of I$_2$.

As a means of experimentally verifying and quantifying the gains associated with the implementation of the external etalon, a preliminary scan across the R56(32,0) iodine transition, utilizing the experimental apparatus shown in Fig. 10 (albeit with the addition
of an external etalon mounted to a 4-axis translation and rotation stage enabling fine angular tuning) was performed. The results of this scan are presented in Fig. 22, along with the results obtained for the same \( I_2 \) cell conditions, but without the etalon.

![Graph showing measured transmission profile over the R56(32,0) transition near 18788.3 cm\(^{-1}\) using the injection-seeded, Q-switched, pulsed laser. Results are shown for scans obtained both with and without the external etalon placed in the optical beampath, along with the transmission profile predicted using the \( I_2 \) spectral model [12].]

These preliminary results are quite encouraging, as the optical density has been increased from a measured value of \(~5\) to \(~7\) for the same cell conditions by introducing the etalon.
This value represents the highest OD reported by more than three orders of magnitude for an FRS diagnostic operating in the visible regime with a pulsed laser source.
In this chapter, two-dimensional imaging of gas-phase fields using FRS within heavily-seeded particle-laden nitrogen and propane jets is demonstrated. This experimental configuration is meant to be used as a somewhat generic test bed for the FRS diagnostic. The experimental setup for the 2D imaging studies is shown in Fig. 23. Frequency-doubled, 532-nm light from a Q-switched, injection-seeded, Nd:YAG passes through a combination of half-waveplate and thin-film polarizer for attenuation of laser energy (if necessary) as well as to ensure vertically-polarized light reaches the experimental test section to maximize the Rayleigh scattering signal. The pulsed-laser output then passes through an externally-mounted etalon. The etalon is mounted on a 4-axis tunable stage which enables fine angle tuning to maximize transmission. Following this, the light passes through an iris to mitigate spatial-frequency fluctuations that may occur in the outer portions of the beam diameter [38] before being focused into a sheet by a single plano-convex cylindrical lens. The 2D laser sheet then passes over a 7.75-mm jet nozzle centered within a 300 mm x 300 mm coflow of air.
Figure 23. Schematic of Experimental Imaging Setup for Filtered Rayleigh Scattering Diagnostics in Turbulent, Seeded and Un-Seeded jets.
Scattered light, both Mie and Rayleigh, is focused through the temperature-controlled, molecular iodine absorption cell and onto a high-quantum-efficiency CCD camera (LaVision Image Intense). The iodine cell is operated under starved-cell conditions at a partial pressure of 1.05 Torr, and a temperature of 341K ± 0.1K. The cell is 254 mm in length and 76 mm in diameter. Output frequency and laser linewidth of the laser source are monitored passively by a High Finesse WS-U wavemeter with a resolution of 5 MHz and an accuracy of 30 MHz. This wavemeter is automatically calibrated every 10 minutes by a single-frequency, frequency-stabilized cw HeNe laser source, coupled with a single-mode fiber to the wavemeter, thereby limiting any long term drift in the measurements.

Filtered Rayleigh Scattering Results

As a demonstration of the imaging characteristics inherent to our system and to determine the feasibility of FRS in heavily-seeded, or highly Mie scattering-dominant, flows (such as spray environments, highly sooting flames, simultaneous PIV/scalar measurements), preliminary FRS images are obtained within a propane jet, seeded with oil droplets as shown in Fig. 24. In this example, the etalon has been removed from the optical path. Figure 24a shows the flowfield imaged when the laser is off-tuned from an absorption feature of I$_2$. The image acquired in Fig. 24a was taken with ~ 1 mJ/pulse. Conversely, Fig. 24b provides an example image with the laser tuned for maximum Mie scattering rejection through the I$_2$ cell; that is, the laser was tuned to the peak of a strong I$_2$ absorption transition near 18788.3 cm$^{-1}$. The image shown in Fig. 24 b was acquired
with ~ 300 mJ/pulse. Note that in this image, the Rayleigh signal arising from the gas-phase propane jet is clearly visible; however, the Mie scattering signal from the oil droplets has not been eliminated entirely. This is due to the limited amount of suppression obtainable with a pulsed Nd:YAG source, as outlined in Chapter 4. From the signal levels of the Mie scattering in this image pair, accounting for the difference in laser energy and camera f-stop, the Mie scattering is calculated as having been reduced by ~10^4. This level of suppression is consistent with previous work [3,33,34].

Figure 24. Initial feasibility of FRS to detect gas-phase concentrations within a field of liquid droplets. a) Mie scattering from a droplet-laden turbulent jet. b) Rayleigh scattering of the gas-phase within the droplet-laden jet. Droplet scattering is reduced by ~ 10^4
While a reduction in unwanted scattering of $\sim 10^4$ is sufficient under certain applications, the results of Fig.24 seem to indicate that this level of scattering suppression would limit a large number of experimental applications to purely qualitative measurements, as it would be difficult to determine whether the signal collected at a given pixel was the result of a Mie or Rayleigh scattering process.

Subsequently, an additional diagnostic was performed, whereby FRS images were acquired within a densely-seeded, turbulent nitrogen jet, issuing into a 0.3 m/s coflow of air. The jet was seeded with droplets comprised of a low-volatility, non-evaporating oil (Canola Oil), with a mean diameter of 0.3 microns. Laser pulse energy for this study was $\sim 350$ mJ/pulse. As a means of quantifying the effectiveness of the FRS methodology in removing “unwanted” scatter from the non-evaporating oil droplets (thereby simulating possible scattering from PIV particles, spray droplets, or soot particles) probability density functions (pdf) of the measured FRS signal on the jet centerline at an $x/d$ of 10 were determined. The raw FRS images were corrected by applying a background subtraction, a sheet correction accounting for spatial variation in laser sheet intensity and pulse-to-pulse laser energy variations, a normalization of the signal from a pure N$_2$ source (a region outside the jet core with an absence of droplets), and a 5x5 median filtering algorithm. Therefore, in this study, a normalized signal of one ($S_{\text{FRS}}/S_{\text{FRS,N2}} = 1$) denotes a pure N$_2$ signal. It is assumed any values higher than one are a result of scattering from the liquid-phase droplets transmitting through the iodine filter cell and onto the detector. Results from this study are presented in Fig. 25.
Figure 25. Probability density function of recorded normalized FRS signals. A normalized signal of 1 indicates that the signal is from N\textsubscript{2} with no scattering from droplets.

In Fig. 25 results are shown for five sets of data, each corresponding to a specific range of laser wavelengths (or frequencies). Each data set corresponds to a range of 0.000025 nm (0.0008 cm\textsuperscript{-1} or 25 MHz). Also included is a pdf of an unseeded (pure N\textsubscript{2}) case. In the absence of any experimental noise or correction imperfections, this would be a delta function at 1. It should be evident from Fig. 25 that even small changes in the laser output wavelength (or frequency) can result in significant changes in the effectiveness of the
FRS diagnostic. It is therefore concluded that the resulting disparities between the various wavelength bins are a result of non-idealized spectral purity. It is also apparent that this particular methodology, of passively monitoring the inherent wavelength fluctuation of the laser and subsequently parsing out selected wavelength ranges for additional analysis, can be an effective method for optimizing the filtered Rayleigh signal.

More important than the wavelength fluctuation effects is the fact that the pdfs of seeded jet cases vary significantly from the baseline, non-seeded case. This indicates that there is a significant amount of Mie scattering signal transmitting through the I$_2$ cell ($S_{FRS}/S_{FRS,N_2} > 1$). Note that the pdfs are the result of scattering signals that has been suppressed by 4-5 orders of magnitude, a decrease already ten times the highest value reported previously within the literature [3,33,34]. For FRS to be viable in environments with high levels of particle/droplet/surface scattering, further suppression of unwanted signals needs to be realized.

FRS Imaging Results Obtained with External Etalon

The 2D imaging experiments described above was repeated with the addition of the etalon placed in the laser beam path as described in chapter 5. Again, the etalon serves as a spectral filter, which, with appropriate tuning and design, can effectively increase the spectral purity of the laser pulse. Again, the FRS signal is recorded as a function of laser frequency. To enable a comparison with the previous case with no etalon, laser pulse energy (after transmission through the etalon), seeding density,
measurement location, and data reduction were matched to the prior conditions. The results of this study are presented in Fig. 26.

Figure 26. Probability density function of normalized FRS signals using an external etalon. A normalized signal of 1 indicates that the signal is from N\textsubscript{2} with no scattering from droplets.

The results shown with the etalon in Fig. 26, as compared to those of Fig. 25, are quite encouraging. Values corresponding to those that would saturate the detector (S\textsubscript{FRS}/S\textsubscript{FRS,N2}...
> 10) are greatly reduced, and in many cases, eliminated entirely. In addition, for all wavelength bins presented, the pdf closely matches that of the baseline, unseeded case for more than one decade of probabilities. (Note that the unseeded profile data was retaken with the etalon in place). Signals deviating from that of the unseeded case occur less than 5% of the time.

This implies that the spectral purity of the laser source is greatly increased as compared to the case without the etalon and sufficient such that the iodine filter can effectively suppress the Mie scattering signal arising from droplets in this sizing range.

As a further example of the improvements associated with the implementation of the etalon, Fig. 27 shows a set of instantaneous FRS images obtained of a densely-seeded, turbulent propane jet. Flow conditions are similar to those of Fig. 24, however, the seeding density has been increased in order to evaluate the additional amount of suppression (a result of increasing the laser spectral purity) enabled with the use of the external etalon. Figure 27 shows seven un-correlated images for cases with and without the etalon in the beam path. Images shown on the left column were acquired without the etalon and are highly dominated by the interfering Mie scattering signal. It is important to note that in these images, the Mie scattered signal has been reduced by 4-5 orders of magnitude with the I$_2$ cell. However, images shown on the right column correspond to images from a data set acquired with the etalon in the beam path. These images show no sign of droplet scattering, only the gas-phase Rayleigh signal is detected.
Figure 27. Instantaneous images of a densely seeded, turbulent propane jet at an $x/d = 10$.
Images on the left were acquired without the external etalon. Images on the right were acquired with the external etalon in the optical path. These images, although preliminary,
clearly demonstrate the feasibility of the FRS methodology with the addition of an external etalon in flows with high levels of scattering interference.

Application to Turbulent Spray Flows

In this section, one new application of FRS is discussed; that is, application of FRS to turbulent spray flows for determination of fuel vapor concentrations and examination of gas-phase mixing processes. For the majority of practical energy-conversion systems, a liquid fuel spray is injected into an oxidizing environment; where the liquid droplets must disperse, evaporate, and the fuel vapor must molecularly mix with the oxidizer prior to chemical reaction. Although complicated by several physical and chemical processes, liquid-fueled combustors are ultimately limited by droplet vaporization and the subsequent gas-phase mixing process. However, the current understanding of gas-phase mixing processes within turbulent sprays is limited and systematic investigations of the gas-phase processes within spray flows, including vapor production and the subsequent vapor-air mixing, has not been studied in detail. This largely stems from the extreme difficulty of accurately measuring vapor-phase concentrations in the presence of droplets. At any region in space, the co-existence of the liquid-phase droplets makes the independent measurement of the gas phase difficult.

Planar laser-induced fluorescence (PLIF) of fluorescent fuels or fuel tracers is an approach that has been applied in spray environments a [43-48]; however there is one major limitation in using PLIF to obtain accurate vapor phase information in turbulent spray flows: the collected signal originates from both the gas and liquid phase
of the droplet. Therefore, the accuracy of any measurement will be predicated on the ability to accurately discriminate between the liquid-phase and vapor-phase fluorescence signal which cannot be spectrally separated. Commonly, during data reduction, a threshold value is set which defines a lower bound to the liquid phase signal \[44,46,48\]. Signals exceeding this value are assumed to originate from liquid droplets and are subsequently filtered out of the acquired image (typically by means of a median filtering algorithm) or removed entirely. However, the signal arising from small droplets, on the order of the size of a single pixel, may fall below the threshold value and be incorrectly and unknowingly attributed to the vapor-phase. A study by Ritchie and Seitzman \[43\] suggests that for liquid-phase acetone fluorescence, drop diameters on the order of tens of microns (depending on saturation conditions) would generate signals of the same magnitude as those arising from the vapor-phase. There are currently on-going efforts to rectify the phase-discrimination problem with additional measurements such as simultaneous phosphorescence and/or Mie scattering (which are presumed to be sensitive only to the liquid phase) \[49,50\]. In this manner, vapor-phase information can be extracted by comparing multiple techniques; that is, a proper combination of the PLIF and phosphorescence/Mie scattering images would lead to an independent vapor-phase measurement. However, the uncertainties in this approach are still quite high and, to date, there are no proven methodologies for completely isolating the vapor-phase information. It is in this manner that a goal of current and future work is the application of FRS to turbulent spray flows to quantitatively determine gas-phase properties. As an example of preliminary work in visualization of turbulent, evaporating spray flows, images of gas-
phase signal obtained from an acetone spray in an N\textsubscript{2} carrier gas are shown in Fig. 28. In these images, the signal detected arises from the evaporation of the liquid acetone droplets into the turbulent nitrogen jet and ambient atmosphere. No evidence of scattering from the liquid phase droplets is apparent in the images. These, admittedly very preliminary results, indicate that the FRS diagnostic can be a viable tool in imaging the gas phase, turbulent dynamics in a spray flow.

Figure 28. Instantaneous images of the gas-phase FRS signal from a turbulent acetone spray jet in an N\textsubscript{2} carrier gas, at an x/d of 10. These images were acquired with the external etalon in the optical path.
Chapter 7: Conclusions and Future Work

The filtered Rayleigh scattering (FRS) diagnostic has been investigated in this thesis with a goal to further understand previously-reported limitations in scattering suppression and to improve current FRS capabilities with the introduction of an external etalon to improve pulsed laser spectral purity. In general, FRS offers the possibility to obtain both qualitative and quantitative information on a variety of flowfield parameters, often under experimental conditions that would preclude the use of other traditional laser based, optical diagnostics [3,6,8,13,29,33].

In previous work reported in the literature, it has been demonstrated that FRS measurements, in conjunction with a Q-switched, injection seeded, pulsed laser source, are limited by the spectral purity of the laser beam. This factor ultimately determines the effectiveness of the suppression of unwanted signal through the molecular filter and limits the applicability and utility of the technique in flows with high levels of unwanted scattering [3,13,33,34]. In this work, modeling results indicate when the spectral purity of a laser source is less than unity, significant reductions in scattering attenuation capabilities can occur. Subsequent experimental data corroborate the results of the modeling, indicating a good understanding of this fundamentally limiting factor.

Unique to the system presented in this thesis is the addition of an external etalon, which was used to increase the spectral purity of the laser output. The addition of the
etalon to the FRS system resulted in an increase in scattering rejection by an additional two orders of magnitude as compared to the FRS system without the etalon. This implementation allowed for the recovery of visually- and statistically-viable gas-phase information to be recovered from flows heavily seeded with liquid droplets. Initial indications imply that FRS can be extended to a variety of situations with high levels of scattering interference including sooting flames, wall-bounded flows, and environments with particle imaging velocimetry measurements. With the etalon, an optical density of $>7$ was obtained, which represents a value three orders of magnitude greater than any previously reported values in the literature.

**Future Work**

Much of the future work regarding the current FRS approach entails ensuring the validity of potential quantitative measurements. This includes continued work on increasing and maintaining the spectral purity of the laser source. A detailed analysis and optimization of the transmission through the external etalon should be performed. This work should include a study of the effects of laser wavelength fluctuations (which can serve to artificially broaden the average laser lineshape), as well as the etalon transmission dependence on temperature fluctuations and variability in tuning angle. Optimization of the etalon transmission ensures high spectral purity, which then results in the greatest amount of scattering suppression (in flows with negligible Doppler shifts). A reduction in the Mie or surface-scattered signal to a level commensurate with or lower
than the noise inherent to the system is a necessary step in obtaining quantifiable information from the transmitted FRS signal. It is important to note that finite limitations in the achievable OD, even as laser spectral purity approaches unity, may preclude application of this diagnostic in certain situations. Laminar flows seeded with non-evaporating droplets provide a reliable test bed for determining the maximum level of droplet loading which may be successfully rejected for a given experimental system.

Further work toward quantification of the transmitted Rayleigh signal includes continued application, validation, and possible expansion of the Tenti S6 (or similar) Rayleigh scattering model for larger fuels \[9,10\] if the application targets realistic configurations. While these models are largely considered among the most accurate in their prediction of the Rayleigh-Brillouin scattering lineshape, there are significant limitations in regard to their applicability to larger fuel molecules. Young and Kattawar \[51\] highlight some of the disparities of the S6 model when it is applied to larger fuels. For example, as discussed in chapter 1, the S6 model was formulated and validated using only monatomic and diatomic molecules. Extension to realistic fuel molecules will necessitate an accurate model of polyatomic molecules. Also, these Rayleigh-Brillouin models have not been developed for multi-component (i.e. fuel vapor – oxidizer) gas mixtures.

While a modification of the Tenti S6 model for use of larger molecules such as hydrocarbon fuels may be possible, it will require accurate values of shear viscosity, heat conductivity, internal degrees of freedom, and bulk viscosity. Data for these values, especially for more complicated molecules, is limited.
An alternative approach (which may also be used to validate modeling results) is to measure the scattering lineshape directly for a range of gases and mixtures under a variety of thermodynamic conditions. For example, a narrow linewidth (cw or pulsed) laser could be used to illuminate a cell filled with a particular gas mixture (note that the liquid fuels of interest in a spray environment may have low vapor pressures so the exact calibration conditions may be challenging) and spectrally resolve the scattering as to experimentally determine the scattering lineshape. One such method is to use a scanning Fabry-Perot Interferometer, which has been used in validation of the initial modeling work performed by Tenti et al. [10]. In this manner, a direct assessment can be made of the accuracy and validity of any changes to the S6 model, and a library of scattering profiles can be experimentally determined for a variety of experimental conditions.

Investigations in regards to FRS accuracy in the presence of droplets/particles under various loadings should be performed. It is expected that for high levels of particle or droplet loading (pertinent to the near field of a spray environment or highly sooting flames) a finite level of laser attenuation will be observed. Characterization of this process will be essential to determining the level of particle/droplet loading acceptable when utilizing the FRS technique.

In our research group, a primary interest is the application of FRS to turbulent spray flows. Though the technique is likely limited to conditions downstream of a nozzle or injector source (far-field conditions), due to limitations in droplet loading and scattering suppression outlined above, information in regards to the dynamics of vaporization and gas-phase mixing in a spray environment would be invaluable. This is,
or course, dependent upon an adequate understanding of the scattering properties of the fuel being used, an accurate model of the transmission profile of the molecular filter being used (particularly the convolution of this profile with the output of the laser source being used), and sufficient experimental signal to noise levels.

Further complications arising from the adaptation of the technique to more realistic spray environments, such as a heated and/or pressurized chamber, will also need to be addressed. While both temperature and pressure broadening will yield a broader Rayleigh-Brillouin lineshape and therefore more signal transmitted to the detector per scatterer, these also represent two additional variables that directly affect the FRS signal. While averaged values of flow properties may be obtained in these conditions using the frequency scanning method outlined in chapter 3, instantaneous measurements, or measurements obtained at one specific frequency (as would be necessitated in optimization of Mie scattering rejection) may require additional diagnostics to be performed concurrently to sufficiently constrain all associated variables.

Analysis of the limitations associated with the FRS methodology has allowed access to gas-phase information in previously un-optically accessible flow fields; showing, explicitly, the feasibility of the FRS technique within highly scattering environments. Extension to environments with high levels of scattering media holds promise due to the relative simplicity of the experimental setup associated with the technique (a single, spectrally-pure laser source, well-characterized filter, and single detector), and the potential to obtain quantitative measurements. With these
considerations and the improvements introduced within this thesis, FRS appears to be a viable technique moving forward in challenging environments.
References


Appendix A: Forkey Code

The following is an amalgamation of the Forkey code, written in Fortran.

```
PROGRAM I2LINES2

C    this program previously called assignlinera2.f
C    4/4/96 changed array limits from 3000 to 50000
C    LAST REVISIONS MADE ON 10/8/93 BY J. FORKEY, MAE,
C    PRINCETON UNIVERSITY.
C    THIS PROGRAM USES THE MOLECULAR CONSTANTS AND FRANCK
C    CONDON FACTORS
C    TABULATED IN THE PUBLICATION "IDENTIFICATION DES
C    TRANSITIONS DU SYSTEME
C    (B-X) DE LA MOLECULE D'IODE ET FACTEURS DE FRANCK-CONDON
C    14000-15600 CM-1"
C    BY S. GERSTENKORN, AND P. LUC (PUBLISHED BY LABORATOIRE
C    AIME - COTTON
C    C.N.R.S. II 91405 ORSAY, FRANCE, 1986).  THE DATA FILE
C    OF FRANCK-CONDON
C    FACTORS (fcfioded) WAS PROVIDED BY P. LUC
C    THE POSITIONS OF ALL OF THE LINES PREDICTED BETWEEN WNLO
C    AND WNHI
C    WAVENUMBERS ARE CALCULATED AND SAVED TO THE FILE FILNAM
C    TRANSITIONS CONSIDERED ARE THOSE THAT HAVE LOWER LEVEL J
C    VALUE - KJ -
C    BETWEEN 0 AND 200 (OR THE LIMIT DETERMINED BY THE UPPER
C    LEVEL DISSOCIATION
C    LIMIT), AND THAT HAVE UPPER LEVEL V - KVU - BETWEEN 0
C    AND 69, AND THAT HAVE
C    LOWER LEVEL V - KVL - BETWEEN 0 AND 19.

INTEGER
IVL,IVU,J,KVU(1:50000),KVL(1:50000),KJ(1:50000),
&        JMAX(0:69),LJ,LJFMAX(0:69),M,NC
DOUBLE PRECISION EU(0:69),BU(0:69),DU(0:69),
&             HU(0:69),LU(0:69),MU(0:69),
&             EL(0:19),BL(0:19),DL(0:19),HL(0:19),
```

& ER(0:69,0:19,0:200),
& EP(0:69,0:19,0:200),
& FACTOR(0:69,0:19,0:8),EXP,
& ENERGY(1:50000),FCF(1:50000),
& OLDEN,CURENTEN,
& WNLO,WNHI,
& GU,EROT,ETOT
REAL DELTAEQQ(1:50000),DELTAC(1:50000)
CHARACTER*1 RORP(50000)
CHARACTER*20 FILNAM

COMMON/FRCOND/FACTOR,LJFMAX

C VARIABLES:
C    IVL=VIBRATIONAL QUANTUM NUMBER OF LOWER ELECTRONIC
C    STATE (X)
C    IVU=VIBRATIONAL QUANTUM NUMBER OF UPPER ELECTRONIC
C    STATE (B)
C    J=ROTATIONAL QUANTUM NUMBER OF LOWER LEVEL
C    KVU(I)=VIBRATIONAL QUANTUM NUMBER OF LOWER ELECTRONIC
C    STATE (X) FOR  
C        ITH TRANSITION
C    KVU(I)=VIBRATIONAL QUANTUM NUMBER OF UPPER ELECTRONIC
C    STATE (B) FOR  
C        ITH TRANSITION
C    KJ(I)=ROTATIONAL QUANTUM NUMBER OF LOWER LEVEL OF ITH
C        TRANSITION
C    JMAX(I)=MAXIMUM J VALUE ALLOWED WITHOUT EXCEEDING THE
C        DISSOCIATION
C    ENERGY OF THE UPPER ELECTRONIC ENERGY LEVEL, FOR UPPER 
C    VIBRATIONAL
C    LEVEL QUANTUM NUMBER I
C    LJ=ROTATIONAL QUANTUM NUMBER DIVIDED BY 25.  USED AS
C    INDEX IN FRANCK-
C    CONDON FACTOR ARRAY LOADED FROM DATA FILE
C    LJFMAX(I)=MAXIMUM LJ VALUE FOR VIBRATIONAL QUANTUM
C    NUMBER I, FOR WHICH
C    THERE IS A FRANCK-CONDON FACTOR VALUE IN THE FCF 
C    DATAFILE
C    M,NC=INDICES USED TO SORT TRANSITION LINES
C    EU(I),BU(I),DU(I),HU(I),LU(I),MU(I)=MOLECULAR
C    CONSTANTS FOR LEVEL IN
C    UPPER ELECTRONIC STATE (B) WITH VIBRATIONAL QUANTUM 
C    NUMBER I
C    EL(I),BL(I),DL(I),HL(I)=MOLECULAR CONSTANTS FOR LEVEL IN
C LOWER ELECTRONIC STATE (X) WITH VIBRATIONAL QUANTUM
NUMBER I
C ER(I,J,K)=ENERGY IN WAVENUMBERS OF R TRANSITION WITH
UPPER LEVEL
C VIBRATIONAL QUANTUM NUMBER I, LOWER LEVEL VIBRATIONAL
QUANTUM
C NUMBER J, AND LOWER LEVEL ROTATIONAL QUANTUM NUMBER K
C EP(I,J,K)=ENERGY IN WAVENUMBERS OF P TRANSITION WITH
UPPER LEVEL
C VIBRATIONAL QUANTUM NUMBER I, LOWER LEVEL VIBRATIONAL
QUANTUM
C NUMBER J, AND LOWER LEVEL ROTATIONAL QUANTUM NUMBER K
C FACTOR(I,J,K)=FRANCK-CONDON FACTOR READ IN FROM DATA
FILE, FOR
C TRANSITION WITH UPPER LEVEL VIBRATIONAL QUANTUM NUMBER
I, LOWER LEVEL
C VIBRATIONAL QUANTUM NUMBER J, AND ROTATIONAL QUANTUM
NUMBER (K*25)
C EXP=VARIABLE USED TO STORE EXPONENT OF CURRENT FCF
WHILE READING IN
C VALUES FROM DATA FILE
C ENERGY(I)=ENERGY, IN WAVENUMBERS OF ITH TRANSITION
C FCF(I)=FRANCK-CONDON FACTOR FOR ITH TRANSITION
C OLDE,CURENTE=ENERGY VARIABLES USED TO SORT
TRANSITION LINES
C WNLO=LOWER LIMIT ON WAVENUMBER
C WNHI=UPPER LIMIT ON WAVENUMBER
C GU=VIBRATIONAL ENERGY OF UPPER LEVEL RELATIVE TO
MINIMUM ENERGY OF THE
C ELECTRONIC B STATE
C EROT=ROTATIONAL ENERGY OF UPPER LEVEL
C ETOT=TOTAL ENERGY OF UPPER LEVEL RELATIVE TO THE
MINIMUM ENERGY OF THE
C ELECTRONIC B STATE
C DELTAEQQ(I)=NUCLEAR ELECTRIC QUADRUPOLE HYPERFINE
STRUCTURE CONSTANT
C FOR THE ITH TRANSITION
C DELTAC(I)=MAGNETIC NUCLEAR SPIN - MOLECULAR ROTATION
HYPERFINE
C STRUCTURE CONSTANT
C RORP(I)=INDICATES WHETHER THE ITH TRANSITION IS AN R
OR P TRANSITION
C FILNAM=FILE NAME OF THE OUTPUT DATA FILE
C
WRITE(6,5)
5 FORMAT('ENTER LOW WAVENUMBER [CM-1]')
READ(5,*)WNLO
WRITE(6,6)
6 FORMAT('ENTER HIGH WAVENUMBER [CM-1]')
READ(5,*)WNHI
WRITE(6,7)
7 FORMAT('ENTER DATA FILE NAME')
READ(5,8)FILNAM
8 FORMAT(A20)

C

C STORE VALUES FOR MOLECULAR CONSTANTS
DATA EL /0.0000, 213.3023, 425.3742, 636.2103,
& 845.8034,
& 1054.1456, 1261.2284, 1467.0428,
1671.5796, 1874.8295,
& 2076.7829, 2277.4300, 2476.7606,
2674.7643, 2871.4302,
& 3066.7470, 3260.7032, 3453.2871,
3644.4874, 3834.2932/
DATA BL /0.3731114098671146, 0.37196700678292829,
& 0.3708161358474350,
& 0.3696585853818199, 0.3684941526048041,
0.3673226380482058,
& 0.3661438399566286, 0.3649575486897959,
0.3637635411248850,
& 0.3625615750588612, 0.3613513836108117,
0.3601326696242798,
& 0.3589051000695991, 0.3576683004462275,
0.3564218491850814,
& 0.3551652720508696, 0.3538980365444274,
0.3526195463050510,
& 0.3513291355128313, 0.3500260632909881/
DATA DL /0.4547547628207550, 0.4572208485138791,
0.4597534338572310,
& 0.4623730064958022, 0.4650910671516871,
0.4679135797367788,
& 0.470843742553093, 0.4738840805962359,
0.4770378589154716,
& 0.4803098171079618, 0.4837062248696054,
0.4872342586490214,
& 0.4909006993891605, 0.4947099513587618,
0.4986613820736552,
<table>
<thead>
<tr>
<th>DATA HL</th>
<th>0.5027459833079082, 0.5069423531948184, 0.511219994177510, 0.5154939634908213, 0.519698766129423/</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0.5556429468721693, 0.5779567669609448, 0.6012238577098949, 0.6256071108097138, 0.6512174418765087, 0.6781490910146517, 0.7065149233796305, 0.7364817297409003, 0.7683055270447346, 0.8023668589770773, 0.8392060965263935, 0.8795587385465209, 0.9243907123195214, 0.9749336741185324, 1.032720309770618, 1.099619635219621, 1.17782297089012, 1.270125873244745/</td>
</tr>
</tbody>
</table>
DATA BU /0.289256971, 0.287738074, 0.286194827, 0.284626516, 0.283032072, 0.281410363, 0.279760289, 0.278080789, 0.276370807, 0.274629252, 0.272854969, 0.271046725, 0.269203212, 0.267323056, 0.265404834, 0.263447094, 0.261448373, 0.259407207, 0.257322139, 0.255191724, 0.253014530, 0.250789133, 0.248514120, 0.246188084, 0.243809635, 0.241377401, 0.238890046, 0.236346284, 0.233744900, 0.231084776, 0.228364917, 0.225584470, 0.222742758, 0.219839290, 0.216873785, 0.213846178, 0.210756623, 0.207605493, 0.204393366, 0.201121013, 0.197789367, 0.194399505, 0.190952609, 0.187449932, 0.183892767, 0.180282407, 0.176620109, 0.172907068, 0.169144381, 0.165333029, 0.161473851, 0.157567537, 0.153614614, 0.149615449, 0.145570257, 0.141479115, 0.137341988, 0.133158785, 0.128929309, 0.124653511, 0.120331372, 0.115963080, 0.111549116, 0.107090361, 0.102588207, 0.0980446757, 0.0934625284, 0.888453519, 0.841976193, 0.795247025, 0.750747024, 0.701022026, 0.672194237, 0.686208169, 0.701022026, 0.716681823, 0.733238172, 0.750747024, 0.769269693, 0.788872567, 0.809626788, 0.831608011, 0.854896312, 0.879576237, 0.905736950, 0.933472447, 0.962881752, 0.994069070, 0.623184071, 0.634498863, 0.646390069, 0.658936697, 0.672194237, 0.686208169, 0.701022026, 0.716681823, 0.733238172, 0.750747024, 0.769269693, 0.788872567, 0.809626788, 0.831608011, 0.854896312, 0.879576237, 0.905736950, 0.933472447, 0.962881752, 0.994069070.
&
0.113886305,
&
0.132170885,
&
0.155123256,
&
0.183653576,
&
0.218773428,
&
0.261854525,
&
0.315160040,
&
0.382660587,
&
0.471148212,
&
0.591744750,
&
0.762122829,
&
0.101080529,

DATA HU /0.236059709, 0.255494386, 0.276699292,
0.299730171,
&
0.413404624,
&
0.574173652,
&
0.804290883,
&
0.113418442,
&
0.160989561,
&
0.230329254,
&
0.332128328,
&
0.481510676,
&
0.700277217,

0.102714384, 0.106222064, 0.109941901,
0.118068229, 0.168632122, 0.175946007,
0.191771415, 0.200317341, 0.209310816,
0.228729448, 0.239206455, 0.250236030,
0.274103892, 0.287032577, 0.300696490,
0.330497240, 0.346792900, 0.364143905,
0.402468213, 0.423708590, 0.446541820,
0.497730397, 0.526515687, 0.557758756,
0.628793001, 0.669261601, 0.713553235,
0.815487852, 0.874242437, 0.939076958,
0.109040280, 0.117905900/
DATA HU /0.236059709, 0.255494386, 0.276699292,
0.299730171,
&
0.324712735, 0.351828987, 0.381305617,
0.448416290, 0.486654557, 0.528454767,
0.624191446, 0.678915959, 0.738788420,
0.875954939, 0.954371438, 0.104020092,
0.123715432, 0.135004496, 0.147390275,
0.175932160, 0.192361681, 0.210436261,
0.252229941, 0.276344336, 0.302896189,
0.364304519, 0.399712054, 0.438665326,
0.528632851, 0.580463410, 0.637491472,
0.769468565, 0.845821533, 0.930224813,
0.102372922, 0.112758280, 0.124327264, 0.1372577828,
0.231418646, 0.258782507, 0.290246517, 0.326577828,
0.368704986, 0.417755631, 0.475104663, 0.542436409,
0.621825688, 0.715844732, 0.827705783, 0.961453506,
0.11222770, 0.131662627, 0.155321297, 0.184323647,
0.220166265, 0.264867717, 0.321190625, 0.301891905,
DATA LU /0.339067474, 0.382234284, 0.431548074, 0.487312647,
0.698953410, 0.549996849, 0.620256031, 0.787179347,
0.112368044, 0.886268189, 0.997813192, 0.886268189,
0.1818043, 0.126602265, 0.142729375, 0.161026496,
0.297251563, 0.205409358, 0.23226556, 0.262682841,
0.488131426, 0.336458409, 0.380889215, 0.431200222,
0.800885008, 0.552523731, 0.625340663, 0.707695536,
0.131689806, 0.906430138, 0.102612626, 0.116210323,
0.219467754, 0.149354233, 0.169566755, 0.192763252,
0.376107648, 0.250311205, 0.286054391, 0.327616067,
0.671072385, 0.432876190, 0.499557954, 0.578145519,
0.125719862, 0.78132017, 0.912556444, 0.106930958,
0.249022455, 0.148323046, 0.175619147, 0.208716555,
0.527140992, 0.298335492, 0.358971029, 0.433931657,
0.121526919, 0.643766649, 0.790669836, 0.977036282,
& 0.152226450, 0.192124993, 0.244446499, 0.313711182, 0.406325388, 0.531474864, 0.702499197, 0.939045055, 0.127050903, 0.174167121/
& DATA MU /0.106365241, 0.118424920, 0.134878319, 0.156148979, 0.182822101, 0.215614715, 0.255362733, 0.303021172, 0.359676671, 0.426572538, 0.505146923, 0.597084729, 0.704383883, 0.829436694, 0.975127447, 0.114494794, 0.134313350, 0.157482286, 0.184624651, 0.216494888, 0.254005129, 0.298256361, 0.350575430, 0.412559046, 0.486126191, 0.573580707, 0.677686275, 0.801756694, 0.949765250, 0.112647822, 0.133761931, 0.159007402, 0.189214641, 0.225388451, 0.268749686, 0.320789002, 0.383336751, 0.458654473, 0.549555387, 0.659563941, 0.793128221, 0.955904190, 0.115513807, 0.140018365, 0.170320603, 0.208014494, 0.255204147, 0.314687716, 0.390214018, 0.486843002, 0.611455583, 0.773479699, 0.985931567, 0.126691964, 0.164183282, 0.214654816, 0.283217039, 0.377209285, 0.507261119, 0.688903221, 0.945038537, 0.130977838, 0.183447759, 0.259737387, 0.371924957, 0.538937621, 0.790944200, 0.117697738, 0.177852696, 0.273453277/
DO 10 IVL = 0,19
   BL(IVL)=BL(IVL)*0.1
   DL(IVL)=DL(IVL)*1.D-8
   HL(IVL)=HL(IVL)*-1.D-15
10 CONTINUE
DO 20 IVU = 0,69
   IF (IVU.LE.64) THEN
      BU(IVU)=BU(IVU)*0.1
   ELSEIF ((IVU.GE.65).AND.(IVU.LE.69)) THEN
      BU(IVU)=BU(IVU)*0.01
   ENDIF
   IF (IVU.LE.19) THEN
      DU(IVU)=DU(IVU)*1.D-8
   ELSEIF ((IVU.GE.20).AND.(IVU.LE.66)) THEN
      DU(IVU)=DU(IVU)*1.D-7
   ELSEIF ((IVU.GE.67).AND.(IVU.LE.69)) THEN
      DU(IVU)=DU(IVU)*1.D-6
   ENDIF
   IF (IVU.LE.17) THEN
      HU(IVU)=HU(IVU)*(-1.D-14)
   ELSEIF ((IVU.GE.18).AND.(IVU.LE.42)) THEN
      HU(IVU)=HU(IVU)*(-1.D-13)
   ELSEIF ((IVU.GE.43).AND.(IVU.LE.62)) THEN
      HU(IVU)=HU(IVU)*(-1.D-12)
   ELSEIF ((IVU.GE.63).AND.(IVU.LE.69)) THEN
      HU(IVU)=HU(IVU)*(-1.D-11)
   ENDIF
   IF (IVU.LE.10) THEN
      LU(IVU)=LU(IVU)*(-1.D-20)
   ELSEIF ((IVU.GE.11).AND.(IVU.LE.28)) THEN
      LU(IVU)=LU(IVU)*(-1.D-19)
   ELSEIF ((IVU.GE.29).AND.(IVU.LE.45)) THEN
      LU(IVU)=LU(IVU)*(-1.D-18)
   ELSEIF ((IVU.GE.46).AND.(IVU.LE.58)) THEN
      LU(IVU)=LU(IVU)*(-1.D-17)
   ELSEIF ((IVU.GE.59).AND.(IVU.LE.67)) THEN
      LU(IVU)=LU(IVU)*(-1.D-16)
   ELSEIF ((IVU.GE.68).AND.(IVU.LE.69)) THEN
      LU(IVU)=LU(IVU)*(-1.D-15)
   ENDIF
   IF (IVU.LE.14) THEN
      MU(IVU)=MU(IVU)*(-1.D-25)
   ELSEIF ((IVU.GE.15).AND.(IVU.LE.28)) THEN
      MU(IVU)=MU(IVU)*(-1.D-24)
   ELSEIF ((IVU.GE.29).AND.(IVU.LE.41)) THEN
      MU(IVU)=MU(IVU)*(-1.D-23)
   ELSEIF ((IVU.GE.42).AND.(IVU.LE.52)) THEN
      MU(IVU)=MU(IVU)*(-1.D-22)
   ELSEIF ((IVU.GE.53).AND.(IVU.LE.60)) THEN
      MU(IVU)=MU(IVU)*(-1.D-21)
ELSEIF ((IVU.GE.61).AND.(IVU.LE.66)) THEN
    MU(IVU)=MU(IVU)*(-1.D-20)
ELSEIF ((IVU.GE.67).AND.(IVU.LE.69)) THEN
    MU(IVU)=MU(IVU)*(-1.D-19)
ENDIF

CONTINUE

READ IN FCF VALUES FROM FILE fcfioded
DO 25 IVU=0,69
  IF (((IVU.GE.0).AND.(IVU.LE.29)).OR.
  &   ((IVU.GE.31).AND.(IVU.LE.41)).OR.
  &   (IVU.EQ.43)) THEN
    LJFMAX(IVU)=8
  ELSEIF ((IVU.EQ.30).OR.(IVU.EQ.42).OR.
  &   (IVU.EQ.44).AND.(IVU.LE.46)).OR.
  &   (IVU.EQ.48).AND.(IVU.LE.50)) THEN
    LJFMAX(IVU)=7
  ELSEIF ((IVU.EQ.47).OR.(IVU.EQ.51).OR.(IVU.EQ.52).OR.
  &   (IVU.EQ.54).OR.(IVU.EQ.55)) THEN
    LJFMAX(IVU)=6
  ELSEIF ((IVU.EQ.53).OR.(IVU.EQ.55).OR.(IVU.EQ.56)) THEN
    LJFMAX(IVU)=5
  ELSEIF ((IVU.GE.59).AND.(IVU.LE.63)) THEN
    LJFMAX(IVU)=4
  ELSEIF ((IVU.EQ.58).OR.(IVU.EQ.64).OR.
  &   (IVU.GE.66).AND.(IVU.LE.68)) THEN
    LJFMAX(IVU)=3
  ELSEIF ((IVU.EQ.65).OR.(IVU.EQ.69)) THEN
    LJFMAX(IVU)=2
ENDIF

CONTINUE

OPEN(UNIT=10,FILE='fcfioded',STATUS='OLD')
DO 40 IVL=0,19
  DO 35 IVU=0,69
    DO 30 LJ=0,LJFMAX(IVU)
      READ(10,27)FACTOR(IVU,IVL,LJ),EXP
      27  FORMAT(44X,F4.4,1X,F3.0)
      FACTOR(IVU,IVL,LJ)=FACTOR(IVU,IVL,LJ)*(10.**(-1.*EXP))
    30  CONTINUE
  35  CONTINUE
  READ(10,*)
25  CONTINUE

READ(10,*)

CONTINUE

READ(10,*)

CONTINUE

READ(10,*)
CONTINUE
CLOSE(10)

C

INITIALIZE ENERGY STORAGE VARIABLES

DO 70 IVL=0,19
   DO 60 IVU=0,69
      DO 50 J=0,200
         ER(IVU,IVL,J)=0.
         EP(IVU,IVL,J)=0.
      50 CONTINUE
   60 CONTINUE
  70 CONTINUE

DO 80 N=1,50000
   ENERGY(N)=0.
   KVU(N)=100
   KVL(N)=100
   KJ(N)=500
   RORP(N)='X'
   FCF(N)=0.
   DELTAEQQ(N)=0.
   DELTAC(N)=0.
80 CONTINUE

C

CALCULATE MAXIMUM J VALUES – LIMITED BY DISOCIATION OF B STATE

DO 100 IVU=0,69
   JMAX(IVU)=INT((-4.375*(FLOAT(IVU)))+375.)
   IF (JMAX(IVU).GT.200) THEN
      JMAX(IVU)=200
   ENDIF
100 CONTINUE

C

M=0
DO 130 IVL=0,19
   DO 120 IVU=0,69
      DO 110 J=0,JMAX(IVU)
         ALPHA = FLOAT(J+1)*FLOAT(J+2)
         BETA = FLOAT(J)*FLOAT(J-1)
         GAMMA = FLOAT(J)*FLOAT(J+1)
         ER(IVU,IVL,J)=EU(IVU)+BU(IVU)*ALPHA-
         DU(IVU)*(ALPHA**2)
         & +HU(IVU)*(ALPHA**3)+LU(IVU)*(ALPHA**4)
         & +MU(IVU)*(ALPHA**5)-EL(IVL)-
         BL(IVL)*GAMMA
110 CONTINUE
120 CONTINUE
130 CONTINUE
& +DL(IVL)*(GAMMA**2) - HL(IVL)*(GAMMA**3)

EP(IVU,IVL,J) = EU(IVU) + BU(IVU) * BETA -

DU(IVU) * (BETA**2)
& + HU(IVU) * (BETA**3) + LU(IVU) * (BETA**4)
& + MU(IVU) * (BETA**5) - EL(IVL) -

BL(IVL) * GAMMA
& +DL(IVL)*(GAMMA**2) - HL(IVL)*(GAMMA**3)

IF ((ER(IVU,IVL,J).GE. WNLO).AND.
& (ER(IVU,IVL,J).LE. WNHI)) THEN

M=M+1
ENERGY(M) = ER(IVU,IVL,J)

KVU(M) = IVU
KVL(M) = IVL
KJ(M) = J
RORP(M) = 'R'
FCF(M) = FCFFUNCTION(IVU,IVL,J,RORP(M))
GU = EU(IVU) - 15661.9409
EROT = BU(IVU) * ALPHA -

DU(IVU) * (ALPHA**2.) + HU(IVU) * (ALPHA**3.)
& +LU(IVU) * (ALPHA**4.) + MU(IVU) * (ALPHA**5.)

ETOT = GU + EROT
DELTAEQQ(M) = 1973.0307 - (0.0209608*GU)
& -((1.44052*EROT)/(4381.212-ETOT))
DELTAEQQ(M) = DELTAEQQ(M) / 30000.
DELTAC(M) = (42993.4/((4381.212-GU)**0.82575)) - 28.7532
DELTAC(M) = DELTAC(M) / 3.E7
ENDIF
& (EP(IVU,IVL,J).LE. WNHI)) THEN

M=M+1
ENERGY(M) = EP(IVU,IVL,J)

KVU(M) = IVU
KVL(M) = IVL
KJ(M) = J
RORP(M) = 'P'
FCF(M) = FCFFUNCTION(IVU,IVL,J,RORP(M))
GU = EU(IVU) - 15661.9409
EROT = BU(IVU) * BETA -

DU(IVU) * (BETA**2.) + HU(IVU) * (BETA**3.)
& +LU(IVU) * (BETA**4.) + MU(IVU) * (BETA**5.)

ETOT = GU + EROT
DELTAEQQ(M) = 1973.0307 - (0.0209608*GU) - (1.44052*EROT)/(4381.212-ETOT))
DELTAEQQ(M) = DELTAEQQ(M)/30000.
DELTAC(M) = (42993.4/((4381.212-GU)**0.82575)) - 28.7532
DELTAC(M) = DELTAC(M)/3.E7
ENDIF
110 CONTINUE
120 CONTINUE
130 CONTINUE
C
C PRINT TO OUTPUT FILE IN ORDER OF LOWEST WAVENUMBER TO HIGHEST WAVENUMBER
OPEN (UNIT=40, FILE=FILNAM)
WRITE(40,*) M
OLDEN=WNLO
CURENTEN=WNHI
NC=0
DO 220 L=1,M
   DO 200 N=1,M
      IF ((ENERGY(N).GT.OLDEN) .AND. (ENERGY(N).LT.CURENTEN)) THEN
         CURENTEN=ENERGY(N)
         NC=N
      ENDIF
200 CONTINUE
WRITE (40,210) ENERGY(NC), KVU(NC), KVL(NC), KJ(NC), RORP(NC), FCF(NC),
   & DELTAEQQ(NC), DELTAC(NC)
210 FORMAT (F10.4,
   ',I2,'
   ',I2,'
   ',I3,'
   ',A1,'
   ',E11.4,'
   ',E13.6,
   &',E13.6)
   OLDEN=ENERGY(NC)
   CURENTEN=WNHI
   NC=0
220 CONTINUE
CLOSE (40)
C
STOP
END

******************************************************************************
***************
FUNCTION FCFFUNCTION(IVU, IVL, J, PORR)
***************
C THIS FUNCTION DETERMINES THE FCF VALUE FOR GIVEN IVU, IVL, AND J VALUES,  
C BY EXTRAPOLATING OR INTERPOLATING FROM VALUES IN THE 
C ARRAY FACTOR WHICH 
C WERE READ IN FROM THE FILE fcfioded IN THE MAIN PROGRAM
C
INTEGER IVU, IVL, J, LJFMAX(0:69), LJ1, LJ2
REAL RJ1, RJ2, RJ3, FUNCTION
DOUBLE PRECISION FACTOR(0:69,0:19,0:8)
CHARACTER*1 PORR

COMMON/FRCOND/FACTOR, LJFMAX

C IF J IS GREATER THAN LJFMAX*25 THEN EXTRAPOLATE. IF J 
C IS LESS THAN 
C LJFMAX*25 THEN INTERPOLATE. 
IF (J.GE.(25*LJFMAX(IVU)) ) THEN 
LJ1=LJFMAX(IVU)-1
LJ2=LJFMAX(IVU)
RJ1=25.*FLOAT(LJFMAX(IVU)-1)
RJ2=25.*FLOAT(LJFMAX(IVU))
RJ3=FLOAT(J)
FUNCTION=FACTOR(IVU, IVL, LJ2)+
&       ((FACTOR(IVU, IVL, LJ2)-FACTOR(IVU, IVL, LJ1))
&        /(RJ2*(RJ2+1.)-RJ1*(RJ1+1.))))*
&       (RJ3*(RJ3+1.)-RJ2*(RJ2+1.))
C IF FCF VALUE CAME OUT NEGATIVE WHEN EXTRAPOLATING, THEN 
USE FCF VALUE
C FOR HIGHEST AVAILABLE J VALUE 
IF (FUNCTION.LE.0.) THEN 
FUNCTION=FACTOR(IVU, IVL, LJ2)
WRITE(*,10) PORR, J, IVU, IVL
10  FORMAT('THE 'A1,I3,'('I2'-I2') LINE HAS BEEN 
ASSIGNED AN FCF VALUE')
WRITE(*,20)(LJ2*25), IVU, IVL
20  FORMAT('EQUAL TO THAT FOR THE LINE R OR P 
',I3,'('I2'-I2') SINCE')
WRITE(*,30)
30  FORMAT('THE VALUE OBTAINED BY EXTRAPOLATION WAS 
LESS THAN ZERO')
WRITE(*,*)
ENDIF
ELSE
LJ1=J/25
LJ2=LJ1+1
RJ1=25.*FLOAT(LJ1)
RJ2=25.*FLOAT(LJ2)
RJ3=FLOAT(J)
FUNCTION=FACTOR(IVU,IVL,LJ1) +
& ((FACTOR(IVU,IVL,LJ2) - FACTOR(IVU,IVL,LJ1))
& / (RJ2*(RJ2+1.) - RJ1*(RJ1+1.)))*
& (RJ3*(RJ3+1.) - RJ1*(RJ1+1.))
ENDIF
C
FCFFUNCTION=FUNCTION
C
RETURN
END

PROGRAM I2SPEC4
C
C THIS PROGRAM PREVIOUSLY CALLED I2SPECMODRA4
C DIFFERENCE BETWEEN VERSION 4 AND VERSION 3 IS THAT
C VERSION 4 ONLY CONSIDERS LINES WITHIN 0.2 CM-1 OF THE
C CURRENT FREQUENCY
C THIS PROGRAM WAS WRITTEN BY J. FORKEY WHILE WORKING WITH
W. LEMPERT AND
C R. MILES AT PRINCETON UNIVERSITY'S MECHANICAL AND
AEROSPACE ENGINEERING
C DEPARTMENT
C MOST RECENT REVISIONS MADE IN APRIL 1996
C
C THIS PROGRAM MODELS THE TRANSMISSION OF AN IODINE CELL,
GIVEN ITS LENGTH,
C TEMPERATURE, AND PRESSURE.
C LINE LOCATIONS, FRANCK-CONDON FACTORS, AND TRANSITION
QUANTUM NUMBERS,
C ARE TAKEN FROM THE FILE FILIN, WHICH IS GENERATED BY THE
PROGRAM
C I2LINES2 (ASSIGNLINERA2) WHICH USES THE SPECTROSCOPIC
CONSTANTS AND FRANCK-
C CONDON FACTORS TABULATED IN THE PUBLICATION
"IDENTIFICATION DES
C TRANSITIONS DU SYSTEME (B-X) DE LA MOLECULE D'IODE ET
FACTEURS DE FRANCK-
C CONDON 14000-15600 CM-1" BY S. GERSTENKORN, AND P. LUC
(PUBLISHED BY
C LABORATOIRE AIME - COTTON C.N.R.S. II 91405 ORSAY,
FRANCE, 1986).
NUMBER DENSITIES ARE CALCULATED USING THE IDEAL GAS LAW, AND A PARTITION
FUNCTION USING ENERGY LEVELS CALCULATED USING CONSTANTS IN GERSTENKORN
AND LUC.
HYPERFINE STRUCTURE (DUE TO 1ST ORDER NUCLEAR ELECTRIC QUADRAPOLE
INTERACTION AND MAGNETIC NUCLEAR SPIN – MOLECULAR ROTATION INTERACTION)
IS MODELED BASED ON KROLL & INNES, J. MOL. SPECT., V36, P295-309,
ON LEVENSON & SCHAWLOW, PHYS. REV. A VOL. 6, (1972) 10-20,
AND WITH THE
HYPERFINE STRUCTURE CONSTANTS CALCULATED IN I2LINES2
(ASSIGNLINESRA2) USING
THE FORMULAS IN GLASER, OPTICS COMM., 54, 335-342 (1985)
THE EXPRESSIONS
FOR ABSORPTION CONSTANT ARE TAKEN FROM STEINFELD'S BOOK, "MOLECULAR
SPECTROSCOPY" AND FROM TELLINGHUISEN, J. QUANT. SPECT. RADIAT. TRANSFER,
AVERAGE ELECTRONIC TRANSITION STRENGTHS ARE TAKEN FROM TELLINGHUISEN,
J. CHEM. PHYS. V76 (1982) 4736-4744, OR FOR FREQUENCIES CLOSE TO DOUBLED
ND:YAG, FROM EXPERIMENTS PERFORMED AT PRINCETON UNIVERSITY.
THIS PROGRAM MODELS EACH HYPERFINE LINE AS A DOPPLER BROADENED LINE,
AND IT ASSUMES THAT THE PEAK OPTICAL DEPTHS OF THE HYPERFINE LINES
ASSOCIATED WITH A GIVEN VIBRATIONAL, ROTATIONAL TRANSITION ARE THE SAME
(Strictly true only for "large J" values).

DOUBLE PRECISION CENTWN(1:50000), CENTHYPWN(1:50000,1:6,1:6),
& TRANS(50000)
REAL TEMP, PRES, RLEN, AETS, CENTABS(1:50000), FCF(1:50000),
& RNUM(1:50000), WIDTHHYPWN(1:50000,1:6,1:6), WNSTART, WNSTOP,
& WNSTEP, DELTAEQQ(1:50000), DELTAC(1:50000)
INTEGER IVL(1:50000), IVH(1:50000), IJ(1:50000), NLINES
CHARACTER*1 PORR(1:50000)
CHARACTER*20 FILIN
COMMON/LINEINFO/CENTWN,FCF,IVL,IVH,IJ,PORR,DELTAEQQ,DELTAC
C
C VARIABLES:
C CENTWN(I)= WAVENUMBER OF CENTER OF VIB., ROT. TRANSITION I
C CENTHYPWN(I,M1,M2)=WAVENUMBER OF CENTER OF HYPERFINE TRANSITION
C ASSOCIATED WITH VIB., ROT. TRANSITION I, AND WITH NUCLEAR M VALUES
C EQUAL TO M1 AND M2.
C WIDTHHYPWN(I,K)=DOPPLER WIDTH, IN WAVENUMBERS, OF HYPERFINE TRANSITION
C ASSOCIATED WITH VIB., ROT. TRANSITION I, AND WITH NUCLEAR M VALUES
C EQUAL TO M1 AND M2
C FCF(I)=FRANK-CONDON FACTOR OF TRANSITION I
C PORR(I)=INDICATES WHETHER VIB., ROT. TRANSITION I IS A P OR AN R LINE
C RNUM(I)=NUMBER OF MOLECULES PER CM3, IN CORRECT LOWER VIB., ROT. STATE
C TO BE EXCITED BY TRANSITION I, DIVIDED BY THE NUMBER OF HYPERFINE TRANSITIONS IN THIS VIB., ROT. LEVEL (I.E. NUMBER OF MOLECULES IN ONE HYPERFINE LEVEL OF THE VIB., ROT. STATE CORRESPONDING TO THIS TRANSITION
C IVL(I)=VIBRATIONAL QUANTUM NUMBER OF LOWER ELECTRONIC STATE (X) FOR VIB., ROT. TRANSITION I
C IVH(I)=VIBRATIONAL QUANTUM NUMBER OF UPPER ELECTRONIC STATE (B) FOR VIB., ROT. TRANSITION I
C IJ(I)=ROTATIONAL QUANTUM NUMBER OF LOWER ELECTRONIC STATE (X) FOR VIB., ROT. TRANSITION I (ROT. QUANT. NUM. OF UPPER STATE IS DETERMINED BY PORR(I))
C CENTABS(I)=ABSORPTION COEFFICIENT (INCLUDING LENGTH FACTOR) AT CENTER OF EACH HYPERFINE TRANSITION WITHIN VIB., ROT., TRANSITION I ( THIS
C IS ASSUMED TO BE THE SAME FOR EACH HYPERFINE TRANSITION ASSOCIATED
C WITH A GIVEN VIB., ROT. TRANSITION)
C TEMP=TEMPERATURE OF CELL (KELVIN)
C PRES=PRESSURE OF CELL (TORR)
C RLEN=LENGTH OF CELL (CM)
C AETS=AVERAGE ELECTRONIC TRANSITION STRENGTH
C WNSTEP=WAVENUMBER RESOLUTION
C WNSTART=STARTING WAVENUMBER
C WNSTOP=ENDING WAVENUMBER
C TRANS(K)=TRANSMISSION AT WAVENUMBER=WNSTOP+K*WNSTEP
C NLINES=NUMBER OF LINES USED IN MODEL
C DELTAEQQ(I)=NUCLEAR ELECTRIC QUADRUPOLE HYPERFINE STRUCTURE CONSTANT
C ASSOCIATED WITH THE VIB., ROT. TRANSITION I
C DELTAC(I)=NUCLEAR SPIN - MOLECULAR ROTATION HYPERFINE STRUCTURE
C CONSTANT
C FILIN=NAME OF DATA FILE GENERATED BY I2LINES2 (ASSIGNLINERA2) WITH
C LINE DATA IN IT
C
CALL GETPARS(TEMP,PRES,RLEN,WNSTART,WNSTOP,WNSTEP)
CALL ASSIGNLINES(FILIN,NLINES)
CALL CALCNUMS(TEMP,PRES,IVL,IJ,RNUM,NLINES)
CALL CALCCENTS(CENTWN,CENTHYPWN,NLINES,DELTAEQQ,DELTAC,IJ)
CALL CALCWIDTHS(CENTHYPWN,TEMP,WIDTHYPWN,RNUM,NLINES,IJ)
CALL GETAETS(((WNSTART+WNSTOP)/2.),AETS)
CALL CALCMAXABS(FCF,AETS,TEMP,RNUM,RLEN,CENTABS,NLINES)
CALL CALCTRANS(WNSTART,WNSTOP,WNSTEP,CENTABS,CENTHYPWN,
               & WIDTHYPWN,IJ,TRANS,NLINES,CENTWN)
CALL DATAOUT(TRANS,WNSTART,WNSTOP,WNSTEP,TEMP,PRES,AETS,RLEN,
               & FILIN)
C
STOP
END

********************************************************************
SUBROUTINE GETPARS(TEMP,PRES,RLEN,WNSTART,WNSTOP,WNSTEP)
C THIS SUBROUTINE GETS INPUT FROM THE KEYBOARD
C
REAL TEMP, PRES, RLEN, WNSTART, WNSTOP, WNSTEP
C
WRITE(6,10)
10 FORMAT('ENTER TEMPERATURE OF CELL [K]')
READ(5,*) TEMP
WRITE(6,20)
20 FORMAT('ENTER PRESSURE OF CELL [TORR]')
READ(5,*) PRES
WRITE(6,30)
30 FORMAT('ENTER LENGTH OF CELL [CM]')
READ(5,*) RLEN
WRITE(6,50)
50 FORMAT('ENTER STARTING WAVE NUMBER [CM-1]')
READ(5,*) WNSTART
WRITE(6,60)
60 FORMAT('ENTER ENDING WAVE NUMBER [CM-1]')
READ(5,*) WNSTOP
WRITE(6,70)
70 FORMAT('ENTER RESOLUTION [CM-1]')
READ(5,*) WNSTEP
C
RETURN
END

***********************************************************
*************
SUBROUTINE ASSIGNLINES(FILIN,IEND)
C THIS SUBROUTINE LOADS DATA INTO VARIABLE ARRAYS FROM THE
FILE FILIN WHICH
C WAS GENERATED BY THE PROGRAM I2LINES2 (ASSIGNLINERA2)
C
DOUBLE PRECISION CENTWN(1:50000)
REAL FCF(1:50000), DELTAEQQ(1:50000), DELTAC(1:50000)
INTEGER IVL(1:50000), IVH(1:50000), IJ(1:50000), IEND
CHARACTER*1 PORR(1:50000)
CHARACTER*20 FILIN
COMMON/LINEINFO/CENTWN, FCF, IVL, IVH, IJ, PORR, DELTAEQQ, DE
LTAC
C
WRITE(6,80)

102
FORMAT('ENTER NAME OF FILE WITH LINE DATA IN IT')
READ(5,90)FILIN
90 FORMAT(A20)
C
OPEN(UNIT=30,FILE=FILIN)
READ(30,*)IEND
DO 10 I=1,IEND

READ(30,15)CENTWN(I),IVH(I),IVL(I),IJ(I),PORR(I),FCF(I),
&       DELTAEQQ(I),DELTAC(I)
15 FORMAT(F10.4,1X,I2,1X,I2,1X,I3,1X,A1,1X,E11.4,1X,E13.6,
&       1X,E13.6)
10 CONTINUE
C
RETURN
END

**********************************************************************
*************************************************************
SUBROUTINE CALCNUMS(TEMP,PRES,IVL,IJ,RNUM,NLINES)
C  THIS SUBROUTINE CALCULATES THE TOTAL NUMBER DENSITY (PER CM3) OF
C  MOLECULES USING THE IDEAL GAS LAW, AND THEN CALCULATES
C  THE DENSITY (PER CM3) OF MOLECULES FOR EACH VIBRATION - ROTATION STATE IN
C  THE LOWER
C  ELECTRONIC LEVEL (X), THAT IS INVOLVED IN THE
C  TRANSITIONS CONSIDERED IN THE
C  PROGRAM. FINALLY, THIS NUMBER IS DIVIDED BY THE NUMBER
C  OF NUCLEAR SPIN
C  STATES (SEE KROLL & INNES) THAT ARE POSSIBLE FOR THE
GIVEN IJ VALUE (ODD
C  IJ - 21, EVEN IJ - 15). THIS ASSUMES THAT THE ENERGY
LEVELS ASSOCIATED WITH THE
C  DIFFERENT NUCLEAR SPIN STATES ALL HAVE APPROXIMATELY THE
SAME ENERGY,
C  SO THAT THE "RELATIVE BOLTZMANN FACTORS" ARE ABOUT 1
C  (I.E. EXP((ENUCLEAR1-ENUCLEAR2)/KT)=1). SPECTROSCOPIC
CONSTANTS
C  ARE TAKEN FROM GERSTENKORN AND LUC'S BOOK.
C
REAL RNUM(1:50000),RNTOT,ENERGYWN,RTDEGEN,SPDEGEN,PTFN
INTEGER IVL(1:50000),IJ(1:50000)
DOUBLE PRECISION EL(0:19), BL(0:19), DL(0:19), HL(0:19)
COMMON/CONSTS/EL, BL, DL, HL

C DATA EL / 0.0000, 213.3023, 425.3742, 636.2103, 845.8034,
& 1054.1456, 1261.2284, 1467.0428, 1671.5796, 1874.8295,
& 2076.7829, 2277.4300, 2476.7606, 2674.7643, 2871.4302,
& 3066.7470, 3260.7032, 3453.2871, 3644.4874, 3834.2932/
DATA BL / 0.3731114098671146, 0.3719670067802829, 0.3708161358474350,
& 0.3696585853818199, 0.3684941526048041, 0.3673226380482058,
& 0.3661438399566286, 0.3649575486897959, 0.3637635411248850,
& 0.3625615750588612, 0.3613513836108117, 0.3601326696242798,
& 0.3589051000695991, 0.3576683004462275, 0.3564218491850814,
& 0.3551652720508696, 0.3538980365444274, 0.3526195463050510,
& 0.3513291355128313, 0.3500260632909881/
DATA DL / 0.4547547628207550, 0.4572208485138791, 0.4597534338572310,
& 0.4623730064958022, 0.4650910671516871, 0.4679135797367788,
& 0.4708437425553093, 0.4738840805962359, 0.4770378589154716,
& 0.4803098171079618, 0.4837062248696054, 0.4872342586490214,
& 0.4909006993891605, 0.4947099513587618, 0.4986613820736552,
& 0.5027459833079082, 0.5069423531948184, 0.511211994177510,
& 0.5154939634908213, 0.519698766129423/
DATA HL / 0.5127518681623231, 0.534032291154361, 0.5556429468721693,
& 0.5779567669609448, 0.6012238577098949, 0.6256071108097138,
& 0.6512174418765087, 0.6781490910146517, 0.7065149233796305,
& 0.7364817297409003, 0.7683055270447346, 0.8023668589797073,
& 0.8392060965263935, 0.8795587385465209,
0.9243907123195214,
& 0.9749336741185324, 1.032720309770618,
1.099619635219621,
& 1.177872297089012, 1.270125873244745/
DO 5 I = 0,19
    BL(I)=BL(I)*0.1
    DL(I)=DL(I)*1.D-8
    HL(I)=HL(I)*-1.D-15
5 CONTINUE
C
RNTOT=9.654E18*PRES/TEMP
PTFN=PARTFCN(TEMP)
DO 10 I=1,NLINES
    GAMMA=(FLOAT(IJ(I)))*(FLOAT(IJ(I)+1))
    ENERGYWN=EL(IVL(I))+(BL(IVL(I))*GAMMA)-
(DL(IVL(I))*(GAMMA**2.)
& + (HL(IVL(I))*(GAMMA**3.))
    RTDEGEN=2.*FLOAT(IJ(I))+1.
    IF (MOD(IJ(I),2).EQ.0) THEN
       SPDEGEN=15.
    ELSE IF (MOD(IJ(I),2).EQ.1) THEN
       SPDEGEN=21.
    ENDIF
    RNUM(I)=RNTOT*SPDEGEN*RTDEGEN
& *EXP(-ENERGYWN/(0.6950*TEMP))/PTFN
C SINCE SPIN STATES OF NUCLEI WERE ALL TAKEN TO BE THE
SAME, THIS RNUM
C IS THE NUMBER DENSITY OF MOLECULES IN A GIVEN
VIBRATIONAL ROTATIONAL
C STATE, WITH ANY NUCLEAR SPIN QUANTUM NUMBERS. THE
PROGRAM, HOWEVER
C CALLS FOR THE NUMBER DENSITY OF MOLECULES WITH SPECIFIC
NUCLEAR SPIN
C QUANTUM NUMBERS WITHIN A GIVEN VIBRATIONAL ROTATIONAL
LEVEL.
C THEREFORE, DIVIDE BY THE NUCLEAR SPIN DEGENERACY.
    RNUM(I)=RNUM(I)/SPDEGEN
10 CONTINUE
C
RETURN
END

***********************************************************
****************
FUNCTION PARTFCN(TEMP)
C THIS FUNCTION CALCULATES THE PARTITION FUNCTION FOR IODINE, BY SUMMING
C UP ALL OF THE TERMS FOR THE X ELECTRONIC STATE, WITH VIBRATIONAL
C QUANTUM NUMBER BETWEEN 0 AND 19, AND ROTATIONAL QUANTUM NUMBER
C BETWEEN 0 AND 200, USING THE CONSTANTS IN GERSTENKORN AND LUC'S BOOK.
C NOTE THAT THE DEGENERACY INCLUDES THE DEGENERACY OF A VIBRATIONAL
C ROTATIONAL STATE DUE TO POSSIBLE NUCLEAR SPIN STATES

INTEGER KV,KJ
REAL TEMP,ENERGYWN,SUMROT,SUM,SPDEG,PARTFCN
DOUBLE PRECISION EL(0:19),BL(0:19),DL(0:19),HL(0:19)
COMMON/CONSTS/EL,BL,DL,HL

C ADD UP TERMS IN PARTITION FUNCTION
SUM=0.
DO 60 KV=0,19
   SUMROT=0.
   DO 50 KJ=0,200
      ENERGYWN=EL(KV)+BL(KV)*FLOAT(KJ)*(FLOAT(KJ)+1.)
      & DL(KV)*(FLOAT(KJ)**2)*((FLOAT(KJ)+1.)**2)
      & +HL(KV)*(FLOAT(KJ)**3)*((FLOAT(KJ)+1.)**3)
      IF (MOD(KJ,2).EQ.0) THEN
         SPDEG=15.
      ELSE IF (MOD(KJ,2).EQ.1) THEN
         SPDEG=21.
      ENDIF
      SUMROT=SUMROT+SPDEG*(2.*FLOAT(KJ)+1.)
      & *EXP(-1.*ENERGYWN/(0.695*TEMP))
   END
   ROTLAST=SPDEG*(2.*FLOAT(KJ)+1.)
   & *EXP(-1.*ENERGYWN/(0.695*TEMP))
   IF ((KJ.EQ.200).AND.((ROTLAST/SUMROT).GT.(.05))) THEN
      WRITE(*,20)KV
      20 FORMAT('THE LAST ROTATIONAL TERM FOR V=',I2,'
      IN THE')
      WRITE(*,30)
   END
60 CONTINUE

106
FORMAT('PARTITION FUNCTION, IS GREATER THAN 5% OF THE SUM')
WRITE(*,40)
FORMAT('OF ALL OF THE OTHER ROTATIONAL TERMS IN THIS')
WRITE(*,45)
FORMAT('VIBRATIONAL LEVEL - MORE ROTATIONAL LEVELS MAY BE')
WRITE(*,48)
FORMAT('NEEDED FOR THIS TEMPERATURE')
ENDIF
50 CONTINUE
SUM=SUM+SUMROT
60 CONTINUE
IF ((SUMROT/SUM).GT.(.05)) THEN
WRITE(*,70)
FORMAT('THE CONTRIBUTION TO THE PARTITION FUNCTION FROM THE TERMS')
WRITE(*,80)
FORMAT('WITH V=19 IS LARGER THAN 5% OF THE SUM OF THE TERMS FROM')
WRITE(*,90)
FORMAT('THE LOWER VIBRATIONAL LEVELS - MORE VIBRATIONAL LEVELS')
WRITE(*,100)
FORMAT('MAY BE NEEDED FOR THIS TEMPERATURE')
ENDIF
PARTFCN=SUM
C
RETURN
END

*********************************************************************************************
*********************************************************************************************

SUBROUTINE CALCCENTS(CENTWN,CENTYPWN,NLINES,DELTAEQQ,DELTAC,IJ)
C  THIS SUBROUTINE CALCULATES THE POSITIONS OF THE HYPERFINE LINES (IN
C  WAVENUMBERS) GIVEN THE POSITIONS OF THE VIBRATION-ROTATION TRANSITIONS.
C  THE NUCLEAR ELECTRIC QUADRUPOLE HYPERFINE INTERACTION AND THE MAGNETIC
C  NUCLEAR SPIN - MOLECULAR ROTATION HYPERFINE INTERACTION ARE INCLUDED.
C THE EQUATIONS USED ARE TAKEN FROM LEVENSON AND SCHAWLOW WHICH ARE,
C STRICTLY SPEAKING VALID ONLY FOR "LARGE J"
C
DOUBLE PRECISION
CENTWN(1:50000),CENTHYPWN(1:50000,1:6,1:6)
REAL DELTAEQQ(1:50000),DELTAC(1:50000),RM1,RM2,RJ
INTEGER NLINES,IJ(1:50000),M1,M2,M2MAX
C
DO 30 I=1,NLINES
   DO 20 M1=1,6
      IF (MOD(IJ(I),2).EQ.0) THEN
         M2MAX=M1-1
      ELSE
         M2MAX=M1
      ENDIF
      DO 10 M2=1,M2MAX
         RM1=FLOAT(M1)-3.5
         RM2=FLOAT(M2)-3.5
         RJ=FLOAT(IJ(I))
         IF (IJ(I).NE.0) THEN
            CENTHYPWN(I,M1,M2)=CENTWN(I)+
            & DELTAC(I)*(RJ*(RM1+RM2)
            & +0.5*(RM1*(RM1+1.)+RM2*(RM2+1.))-8.75)
            & +((DELTAEQQ(I)/80.)*(3.*(RM1**2.+RM2**2.)
            & +(3./RJ)*(RM1*(RM1*(RM1+1.)-
            8.25)+RM2*(RM2*(RM2+1.)-8.25))
            & -17.5))
         ELSE
            CENTHYPWN(I,M1,M2)=CENTWN(I)
         ENDIF
      CONTINUE
   CONTINUE
  CONTINUE
C
RETURN
END

**********************************************************************
*********************
SUBROUTINE
CALCWIDTHS(CENTHYPWN,TEMP,WIDTHHYPWN,RNUM,NLINES,IJ)

108
C THIS SUBROUTINE CALCULATES THE HALF WIDTH AT 1/e MAX FOR EACH OF THE
C HYPERFINE LINES
C
DOUBLE PRECISION CENTHYPWN(1:50000,1:6,1:6)
REAL RNUM(1:50000), WIDTHHYPWN(1:50000,1:6,1:6)
INTEGER IJ(1:50000),M1,M2,M2MAX
C
DO 30 I=1,NLINES
   DO 20 M1=1,6
      IF (MOD(IJ(I),2).EQ.0) THEN
         M2MAX=M1-1
      ELSE
         M2MAX=M1
      ENDIF
      DO 10 M2=1,M2MAX
         WIDTHHYPWN(I,M1,M2)=(2.700E-8)*CENTHYPWN(I,M1,M2)*(SQRT(TEMP))
      10 CONTINUE
   20 CONTINUE
30 CONTINUE
C
RETURN
END

*****************************************************************
*************************
SUBROUTINE GETAETS(WNUM,AETS)
C THIS SUBROUTINE DETERMINES THE AVERAGE ELECTRONIC
C TRANSITION STRENGTH
C OF THE B TO X TRANSITION TO BE USED IN THE CALCULATION
C OF CELL
C TRANSMISSION. THIS SUBROUTINE USES EITHER THE VALUE FOR
C AETS TABULATED IN
C TELLINGHUISEN (1982) (AFTER USING LINEAR INTERPOLATION),
C OR IT USES THE VALUE
C OF 0.99E-36 WHICH WAS DETERMINED AT PRINCETON FOR THE
C FREQUENCY DOUBLED
C ND:YAG WAVELENGTH REGION (18787CM-1 TO 18789CM-1). THIS
C VALUE IS BASED ON 57
C MEASURED VALUES AND IS PROBABLY MORE ACCURATE THAN THE
C VALUE OF
C 1.06E-36 GIVEN BY TELLINGHUISEN (1982) FOR THIS
C WAVELENGTH REGION, AND FOR
THE FCFS USED IN THIS PROGRAM.

DIMENSION STRENGTH(45:63)
REAL WNUM,AETS,WAVE
INTEGER I,J

DATA STRENGTH /0.56, 0.63, 0.67, 0.72, 0.78, 0.83,
0.92, 0.94,
& 1.06, 1.05, 1.02, 1.06, 1.01, 1.01,
1.01, 1.23,
& 1.07, 1.27, 0.95/

WRITE(6,10)
10 FORMAT('DO YOU WANT TO USE PUBLISHED VALUES FOR THE AVERAGE')
WRITE(6,11)
11 FORMAT('ELECTRONIC TRANSITION STRENGTH, OR THE VALUE DETERMINED')
WRITE(6,12)
12 FORMAT('EXPERIMENTALLY AT PRINCETON FOR FREQUENCY DOUBLED ND:YAG?')
WRITE(6,13)
13 FORMAT('TYPE 1 TO USE PUBLISHED VALUES (FOR ALL WAVENUMBERS OTHER
& THAN THOSE')
WRITE(6,14)
14 FORMAT('CLOSE TO 18788CM-1')
WRITE(6,15)
15 FORMAT('TYPE 2 TO USE PRINCETON VALUE (FOR FREQUENCY DOUBLED ND:YAG
& I.E. CLOSE ')
WRITE(6,16)
16 FORMAT('TO 18788CM-1')
READ(5,20)J
20 FORMAT(I1)

IF (J.EQ.1) THEN
  WAVE=1.0E7/WNUM
  I=NINT(WAVE/10)
  AETS=STRENGTH(I)+((WAVE/10.-FLOAT(I))*(STRENGTH(I+1)-STRENGTH(I)))
  AETS=AETS*1.E-36
ELSEIF (J.EQ.2) THEN
  AETS=0.99E-36
ELSE
  AETS=0.97E-36

110
C AETS=0.94E-36
ELSE
  GOTO 5
ENDIF
C
RETURN
END

*************************************************************************
*****************************
SUBROUTINE CALCMAXABS(FCF,AETS,TEMP,RNUM,RLEN,CENTABS,NLINES)
C THIS SUBROUTINE CALCULATES THE PEAK OPTICAL DEPTH AT LINECENTER FOR
C EACH VIB-ROT LINE (THIS IS ASSUMED TO BE THE SAME FOR ALL HYPERFINE
C LINES ASSOCIATED WITH A GIVEN VIB-ROT LINE).
C
REAL FCF(1:50000),RNUM(1:50000),AETS,TEMP,RLEN,CENTABS(1:50000)
C
DO 20 I=1,NLINES
  CENTABS(I)=RLEN*(RNUM(I)/(SQRT(TEMP)))*(4.348E24)*AETS*FCF(I)
20  CONTINUE
C
RETURN
END

*************************************************************************
*****************************
SUBROUTINE CALCTRANS(WNSTART,WNSTOP,WNSTEP,CENTABS,CENTHYPWN, &
WIDTHHYPWN,IJ,TRANS,NLINES,CENTWN)
C THIS SUBROUTINE CALCULATES THE TRANSMISSION OF THE IODINE CELL (NOT
C INCLUDING WINDOW ABSORPTION, REFLECTION ETC.), FOR WNSTART WAVE NUMBERS
C TO WNSTOP WAVE NUMBERS, AT INTERVALS OF WNSTEP WAVE NUMBERS.
C
DOUBLE PRECISION CENHYPWN(1:50000,1:6,1:6), WAVENUM, & TRANS(500000), CENTWN(1:50000)
REAL WNSTART, WNSTOP, WNSTEP, CENTABS(1:50000), & WIDTHHYPWN(1:50000,1:6,1:6)
INTEGER IJ(1:50000), M1, M2, M2MAX, ILO, IHI

C
DO 10 M=1,500000
   TRANS(M)=1.
10 CONTINUE
ILO=1
IHI=2
NPTS=NINT((WNSTOP-WNSTART)/WNSTEP)
DO 130 N=1,NPTS
   WAVENUM=DBLE(WNSTART)+DBLE(FLOAT(N))*DBLE(WNSTEP)
   IF (DMOD(WAVENUM,DBLE(1.0)).LT.WNSTEP) THEN
      WRITE(6,20)WAVENUM
20 FORMAT ('WORKING ON 'F11.4' CM-1')
   ENDIF
50 IF ((WAVENUM-CENTWN(ILO)).GE.(0.2)) THEN
      ILO=ILO+1
      GOTO 50
   ENDIF
60 IF (((CENTWN(IHI+1)-WAVENUM).LE.(0.2)).AND. & (IHI.NE.NLINES)) THEN
      IHI=IHI+1
      GOTO 60
   ENDIF
DO 120 I=ILO,IHI
   DO 110 M1=1,6
      IF (MOD(IJ(I),2).EQ.0) THEN
         M2MAX=M1-1
      ELSE
         M2MAX=M1
      ENDIF
   DO 100 M2=1,M2MAX
      TRANS(N)=TRANS(N)*EXP(-1.*CENTABS(I)* & EXP(-1.*(((WAVENUM-CENTHYPWN(I,M1,M2))/ & WIDTHHYPWN(I,M1,M2))**2.)))
100 CONTINUE
110 CONTINUE
120 CONTINUE
130 CONTINUE
C
RETURN
SUBROUTINE DATAOUT(TRANS, WNSTART, WNSTOP, WNSTEP, TEMP, PRES, AETS, 
& RLEN, FILIN)
C THIS SUBROUTINE SENDS THE DATA TO AN OUTPUT FILE
C
DOUBLE PRECISION WAVENUM, TRANS(500000)
REAL WNSTART, WNSTOP, WNSTEP, TEMP, PRES, AETS, RLEN
CHARACTER*20 FILNAM, FILIN
CHARACTER*30 HEADER
C
HEADER = '# OUTPUT FROM I2SPEC4 #'
WRITE(6, 10)
10 FORMAT('ENTER OUTPUT DATA FILE NAME')
READ(5, 15) FILNAM
15 FORMAT(A20)
OPEN(UNIT=40, FILE=FILNAM)
WRITE(40, 20) HEADER
20 FORMAT(A30)
WRITE(40, 21) FILIN
21 FORMAT('# LINE POSITIONS TAKEN FROM FILE ', A20, ' #')
WRITE(40, 22) TEMP
22 FORMAT('# TEMP = ', F10.5 ' K #')
WRITE(40, 24) PRES
24 FORMAT('# PRES = ', F10.5 ' TORR #')
WRITE(40, 25) RLEN
25 FORMAT('# CELL LENGTH = ', F10.5 ' CM #')
WRITE(40, 26) AETS
26 FORMAT('# AETS = ', E15.5 '[ESU^2 CM^2] #')
WRITE(40, 27) WNSTART, WNSTOP
27 FORMAT('# STARTING AND ENDING WAVENUMBERS ARE 'F15.5,1X,F15.5,' #')
WRITE(40, 28) WNSTEP
28 FORMAT('# RESOLUTION IS ', F10.5 ' #')
NPTS = NINT((WNSTOP - WNSTART) / WNSTEP)
DO 30 N = 1, NPTS
   WAVENUM = DBLE(WNSTART) + DBLE(FLOAT(N)) * DBLE(WNSTEP)
   WRITE(40, 29) WAVENUM, TRANS(N)
30 CONTINUE
CLOSE (40)
Lineshape information for other lines and factors referenced in the code are available by request.