Mole Fraction Measurement Through a Transparent Quarl Burner Using Filtered Rayleigh Scattering

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A filtered Rayleigh scattering system is developed and applied to measure the mole fraction of methane in a methane-air swirl flow through a transparent conical quartz quarl. Light scattering from the location where the laser beam is incident on the surface of the quarl is orders of magnitudes larger than Rayleigh scattering from the gas mixture of interest. This diffusive scattering is suppressed using molecular absorption by an iodine cell and using spatial filtering by an optical aperture. Residual stray light accounted for up to 5% of the total signal and had to be removed for accurate measurements. The flow consisted of a non-premixed mixture of methane and air in the central jet surrounded by a strong swirling air flow. Measurements were conducted at a height of 4 mm from the fuel tube's exit for six different conditions of the swirl flow to demonstrate the ability of the instrument to study the effects of swirl strength and fuel flow rate on the mixing process. By using a 4-leg pulse stretcher to allow higher laser energies in the probe volume, large collection optics and a reference iodine cell to monitor laser wavelength variations, standard deviations of ~ 0.006 in air and ~ 0.012 in a laminar methane flow were achieved for mole fraction measurements.

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1. INTRODUCTION

Application of laser diagnostics to canonical jet flames have greatly contributed to the advancement of combustion science [1, 2]. Detailed measurements of temperature, species and velocities in canonical, non-premixed flames using various diagnostic techniques have provided valuable datasets for validation of turbulence-chemistry interaction models employed in large eddy simulations (LES). Swirl stabilized flames, relevant to gas turbine applications, present a challenging test case for LES models because of the complex coupling between velocity flow field, mixing field, and heat release [3, 4]. Although several experimental datasets are available for open-jet swirl stabilized flames [5-7], data collected inside a conical quarl are rare because of the elevated technical difficulty of such measurements. The use of a quarl and a swirl flow are common in the design of modern gas turbines and industrial boilers to improve flame stability and increase fuel efficiency [8-10]. The effect of quarl in increasing the blow-out limit of swirl stabilized non-premixed flames have been investigated in recent years [4]. Elbaz et al. conducted temperature measurements using a fine-wire thermocouple as well as qualitative OH-PLIF measurements, but no quantitative species measurements inside the quarl are available [3, 11]. Given the importance of mixing in non-premixed flames, experimental measurements of species mole fractions inside the conical quarl are highly desirable. The primary goal of this work is the development of an instrument for mole fraction measurements in non-

reacting confined flows that are suitable for validation of LES models. Non-reacting flow measurements decouple the effects of turbulent mixing from that of heat release, providing further insight into the mixing process and validation datasets to ensure that the simulations fully capture the mixing process [12]. LES validation datasets require spatially ($\sim 100 \mu\text{m}$) and temporally ($< 1 \mu\text{s}$) resolved measurements. These requirements rule out continuous-wave laser-based diagnostics, whether spatially or temporally unresolved. High-accuracy and precision are desirable, and therefore 1D measurements are preferred.

Coherent anti-Stokes Raman spectroscopy (CARS) is often used for temperature and species concentration measurements in combustion in enclosed flows, but it is a point measurement technique in its conventional form with a spatial resolution close to 1 mm [13]. Recently fs-ps 1D CARS has been demonstrated with good spatial resolution ($< 100 \mu\text{m}$) in the proximity of a wall [14] and in particle-laden flames [15]. The high complexity and cost of a 1D fs-ps CARS system make it prohibitive for most combustion laboratories. 1D Raman-Rayleigh scattering has been widely used to study canonical jet flames providing simultaneous measurements of temperature and major species along a line, with a spatial resolution of $\sim 100 \mu\text{m}$ and temporal resolution of $\sim 400 \text{ ns}$ [16, 17]. The combination of Raman and Rayleigh is needed when both temperature and composition is unknown, but either Raman or Rayleigh scattering alone are sufficient for mixture fraction measurements in a non-reacting binary mixture. However, the peculiar geometry of the quarl makes the implementation of Raman or Rayleigh scattering challenging. High-precision Raman scattering measurements

of fuel/air ratio at atmospheric pressures requires energies of several hundreds of mJ. Optical breakdown and quartz damage threshold limit the maximum allowable energy to a few mJ, unless a long-pulse laser or a combination of pulse-stretcher and multiple lasers is employed, quickly raising the cost and the complexity of the instrument. Rayleigh scattering is orders of magnitude stronger than Raman scattering and good signal to noise ratio can be achieved with modest energy per pulse. In an isothermal binary mixture, the amplitude of the Rayleigh signal is directly proportional to the fuel/air ratio [18], so the technique can be directly employed for mixture fraction measurements. However conventional Rayleigh scattering is not suitable for enclosed combustors since the signal is at the same wavelength of the incident light and the Rayleigh measurements get affected by surface scattering. In the current application, the close proximity of the quartz quarl to the measurement volume leads to a surface scattering contribution that is much larger than the molecular Rayleigh scattering, as shown in Fig. 1. In addition to diffuse scattered light, four very bright spots are visible on the surface of the quarl, caused by multiple reflections of the incident beam on the surface. The luminosity from this region would easily saturate the camera, and needs to be spatially filtered. Two approaches are typically employed to suppress surface scattering. Structured laser illumination planar imaging (SLIPI) and absorption by a molecular filter as in filtered Rayleigh scattering (FRS). Recently SLIPI has become a popular strategy to suppress stray light from surface reflection and Mie scattering [19, 20]. In SLIPI, the laser light is modulated to have structural imprint on its spatial profile and the Rayleigh scattering signal is extracted using lock-in detection or equivalent strategies. However, SLIPI in its current form can only be applied to two-dimensional measurements. In the configuration of interest in this work, the intensity of the stray light is several orders stronger than the molecular scattering, resulting in a reduced dynamic range of the technique. In addition, single-shot measurements using SLIPI are made at the cost of reduced spatial resolution [21].

Fig. 1. Drawing of the conical quarl indicating the locations probed by the laser beam (green ellipses) and a photo of the quarl attached on the burner.

In filtered Rayleigh scattering, the excitation source is an injection-seeded laser which is wavelength tuned to match an absorption line of a molecular filter placed in the collection path of the scattered signal. Mie Scattering from soot or dust particles and specular reflections from surfaces have the same wavelength and spectral bandwidth as of the incident laser light and are strongly absorbed by the molecular filter [22]. Since the Rayleigh scattered light from gas molecules is Doppler broadened due to the random motion of molecules, a significant portion of it can fall outside the absorption profile of the molecular filter and is transmitted through the molecular filter. While mercury [23] absorbs strongly in the wavelength of titanium sapphire laser (~ 254 nm), iodine [24] has strong absorption lines in the proximity of 532 nm, the second harmonic of Nd:YAG lasers. Given the

widespread use of Nd:YAG lasers, iodine cell has been the most common choice for filtered Rayleigh scattering, despite the lower absorption of the iodine compared to mercury. FRS has been extensively used in combustion as well as fluid mechanics [25-27] and has been successfully demonstrated in sprays [28], sooting flames [29] as well as enclosed combustors [30]. In combustion diagnostics, the technique has been used prevalently for 2D temperature measurements, and very few works in the past concentrated on the measurement of mole fraction using FRS. Ground et al. [31] used FRS to obtain a 2D mapping of helium to air ratio in merging and non-merging streamwise vortices in supersonic flows. The focus was on 2D average flow field, and the 20% single-shot uncertainty was deemed acceptable. Allison et al. [28] employed FRS for 2D quantitative measurements of fuel vapor to air in an evaporating spray flow. In their experiment interfering stray light was generated by liquid fuel droplets rather than surface reflections.

In this paper, we develop a novel 1D FRS instrument, for high-precision measurements of methane/air mole fraction ratio in non-reacting swirl flows inside a conical quarl. A combination of molecular filtering, spatial filtering and residual stray light correction are employed to suppress the intense stray light contribution. Strategies to maximize the Rayleigh signal level include the use of a pulse-stretcher to increase the laser energy delivered to the measurement volume, large collection optics, and a high-quantum-efficiency CCD camera. Further details of the optical configuration are provided in Section 3. Careful accounting of laser energy and wavelength variations needed to achieve the reported precision and accuracy are detailed in Section 4. Experimental results for different test cases in the burner are presented in Section 5 to show the capability of the instrument to provide validation datasets over a wide range of conditions. Mean, standard deviations and probability distribution function plots are presented to highlight the need for precise single-shot measurements in studying mixing in these complex flows.

2. DESCRIPTION OF CONICAL QUARL BURNER

The quarl burner used in this study is designed to produce a swirl stabilized, turbulent non-premixed flame. It consists of two concentric tubes: the central tube delivers the fuel and has an inner diameter of 4.4 mm and an outer diameter of 6.3 mm. The fuel in this experiment was methane. The outer tube, with an inner diameter of 27 mm, delivers air. The swirl is generated by high-velocity air flow through four tangential inlets. A diverging conical quarl made of quartz, so that it is optically accessible, was used in the experiment. The half cone angle of the quarl was 15 degrees and the height was 40 mm. The thickness of the walls of the quarl was 2 mm. Fig. 1 shows the schematic diagram of the quarl and also a representative photo of the quarl taken on a DSLR camera with a continuous-wave laser of power < 5 mW and exposure time of 1 second. The near-exit region, inaccessible to conventional Rayleigh scattering, is of great importance when comparing experimental results to numerical simulation. Therefore measurements are conducted at a height of 4 mm from the exit of the fuel tube, as indicated by the green ellipses in the diagram.

Particle image velocimetry (PIV) and planar laser-induced fluorescence (PLIF) imaging of OH have previously been conducted in the flow [3]. While OH PLIF imaging could be conducted for the flow inside the quarl, because the OH fluorescence signal is shifted in wavelength compared to its excitation wavelength, PIV could only be conducted downstream of the quarl's exit due to strong specular scattering from the quarl surface. The initial results in the quarl burner with a swirl flow showed that the burner is a useful test-case for studying turbulence-chemistry interaction in configurations of interest for the design of new gas-turbine combustors.

Fig. 2. The schematic diagram of the experimental configuration for filtered Rayleigh scattering. The green lines represent the laser beam path.

3. OPTICAL CONFIGURATION

Various parts of the optical configuration will be explained in this section. The schematic diagram of the FRS system is shown in Fig. 2. The optical configuration broadly consists of four parts: a laser system, a pulse stretcher, a wavelength variation monitoring unit, and a signal collection system.

FRS requires a narrow-bandwidth laser that is tunable in wavelength so that it can be matched to a strong absorption line of a molecular filter. A Continuum Powerlite injection-seeded Nd: YAG laser was used in our system. A combination of half wave plate and a polarizing beam splitter was used to adjust the polarization of the laser output to the required polarization. Recently, Sutton et al. [32] showed that stray light suppression could be further improved by using a Fabry-Perot (FP) etalon in the beam path to increase the spectral purity of the injection-seeded laser. In this work, we used an air-spaced etalon with a free spectral range of 30 GHz and an effective finesse of 24 at 532 nm, mounted on a 2 axis rotation stage for fine tuning. FP etalon helped to improve the attenuation by the iodine cell by an order of magnitude. An intrinsic benefit of using an etalon is that, when the laser is not properly seeded, the transmission of the etalon drops dramatically, reducing the laser energy delivered to the measurement volume. This reduces the risk of damaging the camera in case the laser becomes unseeded.

The iodine cell (ISSI I2S-10) used in the collection system was 25 cm long and had a clear aperture of 72 mm. The cell is a permanently sealed starved cell, with iodine in the cell as superheated vapor at a set density. This design ensures a stable absorption spectrum. The iodine cell temperature was maintained at 65 °C using an active temperature controller. A preliminary experiment was conducted to characterize the absorption characteristics of the iodine cell. A small amount of laser light was split into two portions. One portion was directed to a diffuser and monitored using a photodiode. The other portion was expanded using a telescope, directed through the iodine cell, refocused towards a diffuser,

and monitored with an additional photodiode. By scanning the seed laser's wavelength by temperature tuning it using the laser software and monitoring the two photodiode signals, we experimentally obtained the transmission and absorption spectrum of the iodine cell near the iodine absorption peak chosen for this experiment. The resolution of the absorption spectra was 0.5 pm, limited by the control of our seed laser. The spectra obtained are shown in Fig. 3. The horizontal axis in the graph represents the laser wavelength relative to the center of the absorption line chosen for the experiment. With the FP etalon in the system, a reduction of ~4000 times was achieved in the transmitted intensity of the laser when its wavelength was tuned to the peak of the absorption profile.

The minimum inner diameter of the quarl was 27 mm, therefore the choice of focusing lens was a compromise between avoiding damage on the quarl surface, and preventing optical breakdown at the measurement volume. A combination of 100 mm and 150 mm focal length lenses resulting in an effective focal length of ~60 mm was chosen for the experiment. This focal length, which is shorter than what is commonly used in Raman/Rayleigh measurement in open-jet flames, reduced the Rayleigh range of the focused beams, limiting the usable region of the measurement volume to 4 mm. In addition, it led to high irradiances at the measurement volume, limiting the energy to only a few millijoules before optical breakdown occurs. The need for higher laser energy while avoiding breakdown is a common issue in techniques such as Raman spectroscopy where the signal is very weak. The strategy to overcome this problem is to use a long-pulse laser as in Ref. [33] or a pulse-stretcher as described in Ref. [34] to stretch the laser pulse in time. A 4-cavity pulse stretcher was used for our set-up.

As shown in Fig. 2, the laser beam was divided into four cavities of different lengths using beam splitters and then recombined. Alignment was aided by a beam profiling camera to ensure proper overlap of the beams and by a photodiode placed near the target region to monitor the evolution of the pulse shape during the experiment. Since each cavity

delays the pulse that passes through it by a different time period, the final combined beam has a broadened pulse duration of ~ 150 ns instead of the 10 ns pulse duration of the original pulse. Thus, the peak intensity reduces considerably, allowing laser energies up to 50 mJ to be sent to the measurement volume, with no optical breakdown in methane. Fig. 4 shows temporal profiles of the laser pulse before and after stretching.

Fig. 3. The absorption and transmission spectra of the iodine cell near the chosen absorption line.

The collection optics design is a major departure from FRS instruments used in previous works. In order to increase the signal-to-noise ratio of the instrument, the collection optics design borrows heavily from 1D Raman spectroscopy instruments, where large collection angles are employed to partially offset the weakness of the technique. The system consists of two achromatic lenses of 100 mm diameter, and 300 mm focal length and two Nikon multi-element lenses of focal length 50 mm (Nikkor AF 50 mm f/1.8D). The 300-mm focal length lens images the Rayleigh signal to an image plane. The first 50-mm focal length Nikon lens then collimates this signal and passes it through the iodine cell. It acts as a telescope by reducing the radius of the collimated Rayleigh signal to that of the Nikon lens diameter (< 50 mm). This helps to minimize the signal loss caused by the iodine cell aperture, because the reduced diameter allows all the signal beam collected by the 50-mm lens to travel through the iodine cell roughly collimated, reducing the vignetting caused by the edges of the iodine cell. The last Nikon lens then focuses the signal on a Princeton Instruments 16-bit electron-multiplied CCD camera operated in low noise mode (CCD mode). This configuration is different from the common FRS set-up where the iodine cell is placed directly between the measurement volume and the single multi-element lens imaging on the camera. Similar placement of iodine cell was previously employed by, Doll et. al. in an endoscopic FRS system [35] for the analysis of ducted gas flows. A 532 nm bandpass filter with a full width at half maximum of 10 nm was placed in front of the camera so that the contribution from the ambient room light outside the 532-nm region was cut-off. Potential sources of stray light contribution were minimized by enclosing the laser beam path and the collection optics with black hardboards, making sure that only light collected by the first achromatic lens reached the camera. The camera was hardware binned 6x6 to increase the signal to noise ratio. The overall magnification of the optical train was 1.1 and the corresponding super-pixel resolution was $\sim 87 \mu\text{m}$, as obtained using a target placed in the object plane. Bright spots caused by the curved surface of the quartz were spatially filtered using a rectangular slit placed at the first image plane. Such a configuration has no effect on the FRS signal collected in the 4-mm usable measurement volume, but it completely removes the bright spots.

Fig. 4. Normalized temporal profiles of the laser pulse before and after stretching.

Shot-to-shot variation in laser energy was monitored using a Coherent energy meter which had a nominal uncertainty of $< 1\%$. A reference iodine cell leg similar to the one used by Kearney et al [26] was used to monitor the shot-to-shot variation in wavelength. For this, the laser light leaking from the back of one of the mirrors was sent through a neutral density filter, a 12.5-cm long iodine cell and a 100 mm plano-convex lens to a photodiode, and the signal was acquired using a high-bandwidth oscilloscope. This reference iodine cell was also maintained at 65°C using a temperature controller. The nominal uncertainty of the photodiode was 3%. The ratio of this transmitted signal to the energy meter reading was used to correct the error caused by laser wavelength jitter, as illustrated in the next section.

4. DATA PROCESSING AND INSTRUMENT CHARACTERIZATION

This section describes the procedure to extract 1D mole fraction profiles of methane from the FRS signal in an air-methane mixture. Strategies to minimize measurement errors are presented, and the precision and accuracy of the instrument are discussed. The filtered Rayleigh scattering signal from a species i transmitted through an iodine cell is given by [36],

$$FRS_i = t_c I_L N \sigma_{R,i} \int_{\nu} \phi_i(P, T, \nu, \theta, \lambda_L, \nu) \phi(P_{cell}, T_{cell}, \nu) d\nu \quad (1)$$

where t_c is a constant that is dependent on the collection system, I_L is the laser intensity, N is the number density of the species, $\sigma_{R,i}$ is the Rayleigh scattering cross section of the species, ν is the frequency, ϕ_i is the lineshape function of the Rayleigh-Brillouin scattering (RBS) signal and ϕ is the transmission profile of the iodine cell. In a pulsed-laser-based system, the laser intensity in the probe volume is determined by the instantaneous laser pulse energy. The lineshape function of the RBS signal is dependent on the pressure P , temperature T , velocity ν , observation angle θ and laser frequency λ_L . The transmission spectrum of the iodine cell is a function of the pressure P_{cell} and temperature T_{cell} inside the iodine cell. Because the cell employs a superheated design that is actively controlled, the transmission spectrum can be assumed to be constant. Therefore, in a stationary isothermal gas sample consisting of a single species at constant pressure, the integral term is only a function of the laser wavelength λ_L .

For the current measurements, we assume a binary mixture of two gases - air and methane - and we neglect the different diffusivity of the various air components. We estimate the FRS signal in the mixture as a linear combination of the FRS signals from air and from methane:

$$FRS = [X_{CH_4} \cdot FRS_{CH_4} + (1 - X_{CH_4}) \cdot FRS_{air}] \quad (2)$$

where X_{CH_4} is the mole fraction of methane. For our conditions, the relation between FRS signal and X_{CH_4} can be assumed to be linear as found in [36] and further validated experimentally in section 4A. The quantities FRS_{CH_4} and FRS_{air} are obtained from calibration in methane and air respectively.

It is more convenient to express the signal from filtered Rayleigh scattering in terms of quantities that can directly be calibrated with our instrument. Let the energy-normalized FRS signal in air, obtained at the reference wavelength, be:

$$k_{air} = FRS_{air}(\lambda_{ref}) \quad (3)$$

Then the air-normalized, energy-normalized FRS signal in species i with the laser at wavelength λ_L , can be defined as:

$$A_i(\lambda_L) = \frac{FRS_i(\lambda_L)}{k_{air}} \quad (4)$$

Combining Eq. 2 and Eq. 4,

$$FRS = [X_{CH_4} \cdot A_{CH_4}(\lambda_L) + (1 - X_{CH_4}) \cdot A_{air}(\lambda_L)] \cdot k_{air} \quad (5)$$

The procedure to calibrate k_{air} , $A_{air}(\lambda_L)$ and $A_{CH_4}(\lambda_L)$ is detailed in Section 4A. In the current configuration, we cannot measure the laser wavelength directly, but we monitor its changes using the reference iodine cell, as shown in Fig. 2. We define a term “wavelength indicator” λ as the ratio of the laser intensity transmitted through the reference iodine cell (photodiode signal) to the laser energy recorded by the energy meter. The wavelength indicator provides a shot-to-shot indication of the wavelength variations. For example, when the laser wavelength is near the maximum-absorption point of the spectrum, the wavelength indicator has its minimum value. As the wavelength moves away from the absorption peak, more light gets transmitted through the iodine cell and the wavelength indicator increases. The mean value of the wavelength indicators over all the measurements in air is chosen as the reference wavelength indicator.

The top image of Fig. 5 shows a typical single-shot image of the filtered Rayleigh scattering signal collected from air inside the quarl, and the bottom image shows a similar image with the wavelength of the laser tuned away from the iodine absorption peak. We split the image along the vertical direction into 3 binning regions as shown by the dotted lines in Fig. 5. We obtained the collected signal C_S by software binning of the central region, since no useful information in the vertical (axis of the burner) direction can be extracted. The top and the bottom regions were used to monitor beam steering effects and to verify that the Rayleigh signal was fully contained in the central bin. For the measurements presented here, beam steering was negligible. The camera signal C_S contains contributions from FRS of the gas mixture, residual stray light S_L originating from the laser beam, and a background contribution B .

$$C_S(n, x, \mathbf{x}_0) = E(n) \cdot t(x, \mathbf{x}_0) \cdot FRS(n, x, \mathbf{x}_0, \lambda) + E(n) \cdot S_L(n, x, \mathbf{x}_0, \lambda) + B(x) \quad (6)$$

where $E(n)$ is the instantaneous laser energy measured using the energy meter, $t(x, \mathbf{x}_0)$ is the peak-normalized optical throughput function of the instrument and $FRS(n, x, \mathbf{x}_0, \lambda)$ is the energy-independent filtered Rayleigh scattering signal. $\mathbf{x}_0$ indicates the position vector of the center of the line measurement in a Cartesian reference system with the origin at the center of the exit of the fuel tube, and z-axis

coincident with the tube axis. The value of $\mathbf{x}_0$ are -2.5 mm, 0 mm and 2.5 mm in our measurements. x indicates the pixel number on the camera, that together with $\mathbf{x}_0$ define the spatial location. The index n identifies the instantaneous measurement at the $(x, \mathbf{x}_0)$ location. These three independent variables n , x and $\mathbf{x}_0$ fully define the signal collected at a specific time and space. The subsections below describe our procedure to estimate the several terms in equations 5 and 6, using a series of calibration data.

Fig. 5. Single-shot filtered Rayleigh scattering signal from ambient air inside the quarl with the laser tuned to the peak of the absorption profile (top) and away from the peak (bottom).

A. Measurements with no quarl

Estimates of $A_{air}(\lambda)$, k_{air} , and of $t(x)$ are obtained from data collected in air, with no quarl, and sufficiently away from the burner exit so that stray light is negligible. Air measurements with no quarl are unaffected by $\mathbf{x}_0$ and $x_{CH_4} = 0$. Then, Eq. 6 reduces to:

$$C_S(n, x) = E(n) \cdot t(x) \cdot A_{air}(\lambda) \cdot k_{air} + B(x) \quad (7)$$

The background contribution $B(x)$ includes the camera offset, and residual illumination not generated by the laser. We measured this contribution $B(x)$ by averaging 100 background images taken with the laser beam blocked. Measurements were repeated at the beginning and the end of the experiment, but no noticeable drift was observed. The experiments were conducted with room lights off; therefore camera offset is the main contribution to the background B . $A_{air}(\lambda)$ accounts for changes in the amount of the Rayleigh-Brillouin spectrum of air transmitted through the iodine cell with varying laser wavelength. Often when implementing FRS for quantitative measurements, data obtained at a wavelength that differs from the optimal wavelength by more than 0.03 pm are rejected [28]. Our system had poor wavelength stability, and this criterion would have led to the rejection of most of our data. Here we describe a procedure to account for wavelength variations, extending the applicability of the technique to instruments like ours, that cannot guarantee high stability of laser wavelength.

The ratio of the instantaneous wavelength indicator to this reference wavelength indicator for the measurements in air at that location is named as “normalized wavelength indicator” λ . The wavelength variation in the laser affects the transmitted Rayleigh-Brillouin signal as well as the stray light contribution, if present (negligible for measurements with no quarl). The blue asterisks (Air 1) in the top image of Fig. 6 shows the peak values of the normalized FRS measurements in air, normalized by the mean peak value, plotted against the normalized wavelength indicators of the corresponding

laser shots. The red circles (Air 2) represent a similar signal in air, but from measurements taken after several hours. Both a wavelength jitter and a wavelength drift can be observed in the plot. A strong correlation between the FRS signal and the wavelength indicator is evident. The orange curve is a quadratic polynomial that provides the best fit to the entire set of data in Fig. 6. The R^2 of the fit is 0.85 for the air measurements. By pooling together the measurements in ambient air taken before and after the measurements in the flow of interest, we assure that it covers the entire range of values of wavelength indicator encountered during the duration of the experiment. Numerical simulation of the FRS signal was conducted to verify this trend by convolving the Rayleigh Brillouin spectrum of air using the Tenti S6 model [36] with the experimentally measured Iodine transmission spectrum and integrating over the wavelength region of interest. The simulation also predicts a similar trend in the FRS signal with wavelength variations that are of the order of one-tenth of a picometer.

Fig. 6. Peak values of FRS signal from two separate measurement sets in air (top) and in methane (bottom) as a function of the wavelength indicator. The orange curve is a second order polynomial fit.

The optical throughput normalization $t(x)$ accounts for variations in the spatial profile of the collected filtered Rayleigh signal caused by changes in the focal spot size along the laser propagation direction and by vignetting effects in the collection optics. The short focal length of the system of lenses used for laser focusing, and the separation between the last two collection lenses necessary to accommodate the 25-cm-long iodine cell, exacerbate this issue and cause an 80% drop of signal intensity 2 mm away from the optical axis. With $A_{air}(\lambda)$ determined in the previous step, we obtain $t(x)$ and k_{air} by rearranging Eq. 7:

$$t(x) \cdot k_{air} = \frac{1}{N} \sum_{n=1}^N \frac{C_s(n, x) - B(x)}{A_{air}(\lambda) E(n)} = t' \quad (8)$$

The peak amplitude of t' is k_{air} , and $t(x)$ accounts for its spatial variation. The red, dashed line in Fig. 7, shows a typical profile for the throughput normalization curve $t(x)$, scaled by a factor of 10.

Fig. 7. Standard deviation in the FRS signal of air as a percentage of the mean FRS signal along the probed region.

Having computed all terms in Eq. 5, we can modify it to extract temporally and spatially resolved measurements of $X_{air}(x, n)$ from the FRS measurements in air. This provides an estimate of the instrument precision and the relative importance of the applied corrections. By neglecting instantaneous energy correction ($E(n)=1$) and wavelength corrections ($A_{air}(\lambda)=1$) the standard deviation in the FRS signal over 100 shots is 6% of its mean value, as indicated by blue asterisks in Fig. 7. The energy correction reduces the standard deviation to 2.4 % (black asterisks), and further adding the wavelength correction it reaches 1.2 % (red asterisks), which is closer to the value reported for conventional Rayleigh scattering. At the edges, the standard deviation is lower (1.6%) because the signal to noise ratio drops. We present results obtained in the flow of interest for a probe volume of ± 1.5 mm around the peak of the normalization curve, where the nominal standard deviation is $\sim 1.3\%$. Further improvement in the correction process could be possible by using a more precise photodiode for wavelength monitoring than the one implemented in the current setup, or by directly measuring the laser wavelength with a wavelength meter as in [27].

Measurements in a pure methane jet close to the exit of the fuel tube provide a similar expression for the wavelength dependence of the methane FRS signal:

$$C_s(n, x) = E(n) \cdot t(x) \cdot A_{CH4}(\lambda) \cdot k_{air} + B(x) \quad (9)$$

The FRS signal vs wavelength indicator curve for methane is also shown in Fig. 6 (bottom). It has a more shallow profile compared to the curve for air, as also predicted by the Tenti S6 model, because of the broader linewidth associated with the lower molecular weight. Including the wavelength correction term $A_{CH4}(\lambda)$ improved the measurement standard deviation from 1.4% to 0.8% in methane. The higher standard deviation for methane data suggests that the precision of the FRS measurements in air was limited by the allowed laser energy.

In order to verify the linearity of FRS signal with mole fraction for our conditions, we performed measurements in mixtures of air and methane, varying the mole fraction of methane from 0 to 1 at a step interval of 0.05. No quartz was used during calibration in order to avoid any uncertainty caused by stray light. The calibration result is shown in

Fig. 8. A similar calibration with no iodine cell was also conducted in order to obtain the conventional Rayleigh scattering measurements. Conventional Rayleigh scattering is linear with mole fraction, and provides an accurate measurement of the methane mole fraction, plotted on the x-axis of Fig. 8, for the calibration measurements. The deviation β from the linear curve is determined by dividing the calibration data with a linear fit to the same data. The plot shows that the FRS behaves very linearly with the Rayleigh signal, for our conditions.

Measurements in helium were also conducted during the experiment, for stray light correction as explained in the next section. The helium FRS cross section was only 3.3% of the air FRS cross section, leading to noisier data. Being a lighter molecule, it is less sensitive to the wavelength shift. Therefore no explicit dependence on the wavelength indicator could be extracted and we make the approximation $A_{He}(\lambda) = A_{He}$.

Fig. 8. Calibration of FRS signal against methane mole fraction in the gas mixture.

B. Measurements in air and helium with the quarl

In this subsection, we discuss the procedure to obtain the FRS signal from air, after inserting the quarl. For air, Eq. 6 can be re-written as

$$C_S(n, x, \mathbf{x}_0) = E(n) \cdot t(x, \mathbf{x}_0) \cdot A_{air}(\lambda) \cdot k_{air} + E(n) \cdot S_L(n, x, \mathbf{x}_0, \lambda) + B(x) \quad (10)$$

The terms $A_{air}(\lambda)$ and $B(x)$ have been obtained in the previous section using the no-quarl case. The throughput normalization curve is now also a function of the spatial coordinate of the measurement volume, $\mathbf{x}_0$. For different values of $\mathbf{x}_0$, the relative position of the measurement volume and the quartz quarl as well as the focusing of the incident beam changes. Surface reflection and absorption due to the quarl, which reduce the signal collected on the camera, are also dependent on the location. Thus, the optical throughput function $t(x, \mathbf{x}_0)$ and k_{air} vary with $\mathbf{x}_0$. Therefore, FRS signal in air is measured separately at each location. The procedure described in the previous section to obtain $t(x, \mathbf{x}_0)$ and k_{air} from measurements in air can be repeated for measurements with the quarl, but the additional term caused by stray light must be taken into account.

It should be noted that in the burner of interest for this study, the stray light signal could be strong enough even to saturate the camera if not attenuated by more than three orders of magnitude by the iodine cell. The bottom of Fig. 5 shows measurements in dry air after detuning the laser wavelength away from the iodine absorption line, allowing more scattered stray light to pass through the iodine cell unabsorbed.

The strategy used in this paper to account for residual stray light (after iodine absorption) is an extension of what is typically used in conventional Rayleigh scattering [37], and involves the use of helium, a weak scatterer, to enhance the contrast between the remaining stray light contribution and the FRS signal contribution. For each location $\mathbf{x}_0$, measurements are taken in air and in helium.

$$C_{He}(x, \mathbf{x}_0, \lambda) = E(n) \cdot [A_{He} \cdot t(x, \mathbf{x}_0) \cdot k_{air} + S_L(x, \mathbf{x}_0, \lambda)] + B(x) \quad (11)$$

$$C_{air}(x, \mathbf{x}_0, \lambda) = E(n) \cdot [A_{air}(\lambda) \cdot t(x, \mathbf{x}_0) \cdot k_{air} + S_L(x, \mathbf{x}_0, \lambda)] + B(x) \quad (12)$$

Indicating the energy-normalized camera signal with C_N

$$C_N = \frac{C - B}{E} = A(\lambda) \cdot t(x, \mathbf{x}_0) \cdot k_{air} + S_L(x, \mathbf{x}_0, \lambda) \quad (13)$$

and the effective filtered Rayleigh cross section of helium, with air as the reference, with α :

$$\alpha(\lambda) = \frac{A_{He}}{A_{air}(\lambda)} \quad (14)$$

where $A_{air}(\lambda)$ and A_{He} are determined from measurements with negligible stray light (no-quarl case), we can express the mean stray light at the location $\mathbf{x}_0$ as:

$$S_L(x, \mathbf{x}_0, \lambda) = \frac{C_{N,He}(x, \mathbf{x}_0, \lambda) - \alpha(\lambda) \cdot C_{N,Air}(x, \mathbf{x}_0, \lambda)}{1 - \alpha(\lambda)} \quad (15)$$

The exact wavelength of the laser might be slightly different between the measurements in helium and the actual experiments in the methane-air flow, due to wavelength jitter and drift. Neglecting stray light dependence on the laser wavelength is acceptable only in close proximity of the iodine absorption peak. In this case, the stray light is attributed prevalently to the lack of spectral purity of the laser and additional laser scattered light that bypasses the iodine cell and reaches the camera. Note the underlying assumption that signal from photons at the iodine absorption peak wavelength is negligible. As the laser wavelength drifts away from the optimal location, the iodine cell becomes less efficient in blocking the light scattered from the quarl, and the contribution of stray light at the laser center wavelength to the FRS signal increases. By its definition, the normalized wavelength indicator using the reference iodine cell directly monitors this process. Then stray light can be expressed as the sum of two contributions:

$$S_L(x, \mathbf{x}_0, \lambda) = S_L(x, \mathbf{x}_0, \lambda_{min}) \cdot (1 + a \cdot (\lambda - \lambda_{min})^2) \quad (16)$$

The quadratic dependence is caused by the fact that the reference iodine cell is only half the length of the one used in the collection system. We sorted the data from the measurements in air with the quarl based on their wavelength indicator and binned in 10 wavelength intervals. Bins containing less than 10 measurements were discarded. $S_L(x, \mathbf{x}_0, \lambda_{min})$ is the stray-light value computed for location $\mathbf{x}_0$ corresponding to the bin region with the lowest λ . In order to determine the coefficient a , we pooled together data corresponding to different pixel locations x and plotted them against λ . Fig. 9 shows this plot for three burner locations, $\mathbf{x}_0 = -2.5$, $\mathbf{x}_0 = 0$ and $\mathbf{x}_0 = 2.5$. We can extract a coefficient $a(\mathbf{x}_0)$ for each separate dataset, or use a single

coefficient a as the best fit to the entire set. Since we could not justify a separate wavelength coefficient based on the location, we decided to use a single coefficient a . We consider the difference between the dashed curve (single fit) and the continuous curves (location dependent fit) as an estimate of the error on the stray light contribution. For the three locations considered here, the wavelength independent stray light contribution accounts for 1 % to 3 % of the signal in air. Within the range of wavelength jitter encountered, the stray light can increase by $50\% \pm 20\%$ leading to contributions up to 4.8% of the signal in air. Neglecting the wavelength dependent stray light contribution would cause over-estimating k_{air} by 1.5% and therefore biasing the methane mole fraction to lower values. Note that, measurements in methane-air mixtures are less sensitive to errors in the stray light correction thanks to the larger Rayleigh cross section of methane.

Fig. 9. Variation in estimated stray light as a function of wavelength indicator, for three different locations.

We validated the approach by evaluating an additional set of measurements in air at $x_0 = 0$ taken after a few hours. There was a 20% increase in the wavelength indicator. Neglecting the wavelength dependence of the stray light, the measured Rayleigh cross section would be overestimated by 1.5%. This analysis also revealed another larger source of inaccuracy. Fig. 10 shows the throughput normalization curves computed independently from the two air measurements. There is a small shift between the profiles. Using the $t(x, x_0)$ computed from the first set of measurements in air (“air 1”) to process data collected later would generate a slope in the measured air concentration, and errors ranging from -10% to +4%. The dashed blue curve is the throughput curve obtained from the “air 1” set shifted by 0.4 superpixels (~ 30 microns on the camera chip). By simply shifting the $t(x, x_0)$ of “air 1”, the discrepancy reduces to $\pm 1.5\%$. The small shift could be attributed to slight changes in the focusing of the laser beam over the several hours of a typical test. This is also a common issue in conventional 1D Rayleigh scattering measurements, and it is typically contained by frequent measurements of the throughput normalization. In our experiment, air measurements were taken only twice, before and after measurements in the flow of interest, and also there is no basis to assume a linear trend of the throughput normalization shift with time. In the experiment presented here, measurements are taken at three locations $(x_0, y) = (-2.5, 4)$, $(x_0, y) = (0, 4)$ and $(x_0, y) = (2.5, 4)$, where y indicates the height from the fuel tube exit. The length of the measurement volume is 3 mm centered at the peak of the throughput curve, providing a 0.5 mm overlap between adjacent measurements. For each flow condition, measurements in the three locations are taken in rapid succession, and it is legitimate to assume a single value of the throughput shift. We then computed the shift as the value that minimized the mismatch in the 2 overlap regions formed by the three

sets of measurements. The residual mismatch is an indication of the measurement inaccuracy after all the corrections have been implemented, and reaches up to 0.035 in mole fraction.

Fig. 10. Throughput normalization function $t(x, x_0)$ obtained from air measurements before and after the experiments, along with the percentage error in their ratio before and after adjustment of shift.

In summary, measurements with no quarl provided the values of $A_{air}(\lambda)$, $A_{CH_4}(\lambda)$, $A_{He}(\lambda)$ and $B(x)$, measurements in air and helium with the quarl attached to the burner provided the values of $S_L(n, x, x_0, \lambda)$, k_{air} and $t(x, x_0)$. For each flow-condition, a sub-pixel shift is added to $t(x, x_0)$ that accounts for small changes in the beam alignment. All the terms in Eq. 6 are known, and then Eq. 5 can be inverted to provide a direct measurement of methane mole fraction which is presented in the next section.

The mole fraction measurements had a standard deviation of 0.006 in air and 0.012 in pure methane. These two values bound the precision of the instrument. This high precision was made possible by the use of pulse stretcher and large collection optics to boost the signal, and by careful accounting of laser wavelength and energy variations. Similar high precision would be difficult to achieve in 2D measurements due to lower laser energy densities and additional shot-to-shot spatial variation in laser sheet profile. The accuracy of the measurements depends strongly on the calibration procedure. Measurements in air and methane with and without quarl were taken immediately before and after measurements in the flow of interest to minimize the bias. One of the assumptions in the current work is that the Doppler shift in the scattered light due to the velocity components of the flow is negligible. The maximum velocity component of the flow in the direction of the laser beam or/and the tangential velocity component would cause a Doppler shift of $\sim \pm 0.014$ pm and the corresponding inaccuracy in the mole fraction would be ± 0.003 . Temperature changes in the mixture for the flow conditions used in this experiment are negligible. Uncertainty in stray light correction caused an additional inaccuracy of 0.008 in mole fraction. Another major source of inaccuracy is the drift in the throughput profile, likely caused by changes in the focusing of the laser beam, resulting in $\sim 1.5\%$ error in the throughput curve corresponding to an inaccuracy of 0.006 in the methane mole fraction. The measured standard deviation in the flow is larger than what we anticipated, and for 500 samples the 95% confidence interval on the sample mean is ~ 0.02 . This is the largest source of inaccuracy in the presented measurements. It should be noted that we consider the worst case scenario in most of these bias estimates (stray light, velocity contribution etc.) and the corresponding average bias would be much lower. As an empirical criterion to estimate the accuracy, the over-all resultant inaccuracy in measurements is estimated as the maximum difference between repeated measurements at the same location from the experiments in the turbulent flame which is 0.035 in mole fraction.

Table 1 Flow conditions considered for the experiment.

	Fuel flow rate (SLPM)	Axial air flow rate (SLPM)	Tangential air flow rate (SLPM)
Condition 1	6	10	190
Condition 2	10	10	190
Condition 3	16	10	190
Condition 4	10	40	160
Condition 5	16	40	260
Condition 6	4	40	120

5. RESULTS AND DISCUSSION

The previous section provided detailed information on how to estimate all terms in Eq. 6, and on the effect of their contributions to the precision of measurements. This section presents the measurement results of methane mole fraction at a height 4 mm from the exit of the fuel tube at radial locations that are 2.5 mm apart along the laser beam direction, as shown in Fig. 1. Six different flow conditions were tested. The mass flow rates for these conditions, expressed in standard liter per minute (SLPM), are tabulated in Table 1.

In the first three conditions, the tangential mass flow rate of air was set to 190 SLPM, the axial flow rate of air was set to 10 SLPM, and the methane flow rate was varied from 6 to 16 SLPM. The fourth test case has the same total air flow rate and methane flow rate as Condition 2, but has a lower swirl number with a tangential air flow rate of 160 SLPM, an axial air flow rate of 40 SLPM, and a methane flow rate of 10 SLPM. Condition 5 has the same axial flow rate as condition 4, but the fuel flow rate is 1.6 times higher and the tangential air flow rate is also increased correspondingly. The 6th Condition is a case with very small fuel and tangential air flow rates. The probe volume length considered for analysis was ~ 3 mm in the laser direction, with a spatial resolution of $\sim 100 \mu\text{m}$, resulting in 30 measurement locations. After this length, the signal dropped to below 30% of that at the beam waist, and the signal-to-noise ratio became poor.

Fig. 11 shows the mean mole fraction of methane in the flow within the quarl, taken over 500 laser shots for all the flow conditions listed in Table 1. The standard deviation of methane mole fraction, indicative of the locations where mixing is occurring, is also shown in the figures (dashed red lines). Fig. 12 shows histograms of methane mole fraction at the centerline and at $x=2.5$ mm (off-axis). Combined with the standard deviation, it provides additional information on the state of mixing. The main observations from these plots are the following:

Fig. 11. Mean and standard deviation of the mole fraction of methane, measured at a height of 4 mm from the nozzle exit for six different conditions.

Fig. 12. Probability-normalized histograms of methane mole fraction at the centerline and at $x=2.5$ mm for six different conditions.

- Conditions 1-3 differ in the mass-flow rate of methane. Results show an increase in the peak methane mole fraction from ~ 0.05 to ~ 0.9 when the mass flow rate of methane increases from 6 SLPM to 16 SLPM. This is expected since a higher flow-rate of methane corresponds to a higher central fuel jet velocity and thus a longer penetration length of the central methane jet.
- In condition 1, the mean methane mole fraction is uniform suggesting effective mixing already at 4 mm. The standard deviation shows that variations are as large as the mean value of 0.05. The probability distribution function (PDF) shows an

approximately bell-shaped distribution centered at this value. PDF at 2.5 mm away from the center, is not significantly different.

- When the methane flow rate is increased to 10 SLPM (Condition 2), an increase in the mean mole fraction to ~ 0.5 is observed along with a high and flat standard deviation of ~ 0.3 throughout the region of investigation. Interestingly the PDF shows a more complex distribution, with peaks for pure air and pure methane, and a wide range of compositions in between. Moving away from the centerline, the standard deviation stays constant, but the mean drops from 0.54 to 0.37. Correspondingly, the off-axis shows a decrease in the peaks near pure methane. The non-Gaussian behavior can be attributed to the center jet precession and the precessing vortex core. [38]
- Further increasing the mole fraction to 16 SLPM (Condition 3), the centerline mole fraction rises to ~ 0.9 and the standard deviation has a minimum on the centerline. The PDF shows that most of the flow constitutes of nearly pure methane. Moving away from the centerline the mole fraction decreases and the standard deviation increases, corresponding to the interface between the inner jet and the surrounding air.
- For the same flow rates of fuel and air, decreasing the swirl weakens the mixing, as can be observed by comparing Conditions 2 which has a swirl number of 14.4 and 4 with Condition 4 which has a swirl number of 10.2.
- Condition 5 has mean, standard deviation and PDF profiles similar to Condition 4. This indicates that, while increasing the fuel flow rate, similar mixing efficiency can be maintained at this height by increasing the tangential velocity correspondingly.
- Condition 6, which has the smallest methane flow rate as well as total air flow rate, shows a mean mole fraction close to 0.5 and a standard deviation of $\sim 30\%$. The PDF shows a wide range of mixing ratios. This shows that a good mixing region is achievable even for a small flow rate of 4 SLPM by reducing the tangential flow velocity accordingly.

These details would be obtained only from single-shot measurements and not from time-averaged mole fraction measurements using continuous wave laser sources. The results highlight the need for temporally and spatially resolved measurements to correctly describe the mixing in swirl flows, and shows that FRS can provide this capability even in challenging geometries with intense scattering.

6. CONCLUSIONS

A novel, one-dimensional filtered Rayleigh scattering system has been developed and applied to measure the mole fraction of methane in a nonreactive methane-air flow through a transparent conical quartz burner. Challenges such as a large amount of stray light and optical distortions introduced by the quartz, and limitations on the maximum allowable irradiance imposed by the damage threshold of the quartz quartz and by optical breakdown in methane were addressed carefully while designing the optical system. The experimental configuration involving a 4-cavity pulse stretcher, FP-etalon and iodine cells, and details of the data analysis process have been presented. Wavelength jitter was found to be responsible for significant uncertainty in the measured signal. A procedure to minimize the effect of jitter using a reference iodine cell was demonstrated. After this correction, a standard deviation of $\sim 1.3\%$ in the FRS signal, close to what is reported for conventional Rayleigh scattering, has been achieved for measurements in air within the quartz. Measurement accuracy is very sensitive to the throughput normalization curve, in particular toward the edges of the measurement volume, because of tight focusing of the beam. The issue was minimized by using only the central portion of the

measurement volume, and taking advantage of the overlap region to improve the measurements accuracy.

Six flow conditions differing in the fuel velocity, axial air velocity or/and tangential air velocity were tested using the FRS system. These conditions were chosen in order to isolate the role of the swirl number and the center jet flow rate on the mixing field. The mean and standard deviation in mole fraction measurements could be successfully used to differentiate these effects. Comparison of the histogram of mole fraction with the mean mole fraction at a given location revealed how mixed the flow was, at that location. These results can serve as important validation test-cases for numerical research groups investigating the corresponding reactive cases. In addition to measuring, for the first time, fuel mole fraction within a conical quarl burner, the objective of this work was to address the suitability of 1D filtered Rayleigh scattering as companion diagnostics to Raman scattering in enclosed combustors. Although the laser used in this work had a poor wavelength stability, we achieved a mole fraction standard deviation of 0.006 in air, and 0.012 in pure methane, after wavelength correction. This suggests that 1D FRS can be coupled to 1D Raman for higher precision measurements, similar to what is done with conventional Rayleigh scattering.

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