High-speed Rayleigh-Raman measurements with subframe burst gating

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A 10 kHz one-dimensional Rayleigh-CH₄ Raman instrument capable of achieving highly precise measurement of temperature and methane mole fraction is demonstrated. The system uses a pulse burst laser as the light source and back-illuminated electron-multiplied CCD cameras as the detectors. The cameras are operated in subframe burst gating mode, to combine high sampling rate, with low-noise, slow-readout. The improved precision of this configuration is demonstrated by measuring temperature and methane mole fraction in ambient temperature gas mixtures and in a non-premixed inverse diffusion flame.

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One-dimensional Rayleigh-Raman scattering is a powerful diagnostic to measure temperature and major species concentration in non-sooting flames with high accuracy and precision, but with sampling rates typically limited to 5 or 10 Hz [1]. The study of combustion dynamics events such as quenching, extinction, and thermo-acoustics instabilities, would greatly benefit from extending the Rayleigh/ Raman diagnostics to kHz rates, while maintaining comparable accuracy and precision. Raman scattering in combustion requires laser pulse energies of the order of 1 J, which is 2 orders of magnitude higher than what is provided by conventional high-speed lasers. Only thanks to the recent development of high-energy pulse-burst lasers [2, 3] that can operate at several kHz and deliver pulses with hundreds of millijoules, Rayleigh and Raman scattering in combustion have been extended to kHz rates [4, 5].

The high precision and accuracy of low-repetition-rate Raman systems are enabled by using low-noise CCD cameras, which are generally limited to an acquisition rate of 5 Hz. Previous pulse-burst Rayleigh-Raman scattering instruments employed high-speed CMOS cameras as detectors due to their high acquisition rate [4, 5]. High-speed CMOS cameras have inferior noise performance, lower dynamic range, and poorer quantum efficiency than back-illuminated CCD cameras, resulting in lower signal-to-noise ratio (SNR) and instrument precision. In particular for pulse-burst Raman spectroscopy the scattered signal is weak, and a high-speed

intensifier is needed, to boost the signal on the CMOS camera while suppressing flame luminosity. The use of high-speed intensifier provides a fast gating that suppresses flame luminosity, but results in poorer precision and loss of spatial resolution. Ajrouche et al. [6] demonstrated that electron multiplying CCD (EMCCD) cameras can provide a higher signal to noise ratio than ICCD camera for weak Raman signal, and are better detectors to extend the detection limit beyond the readout noise limit. An external shutter that consists of a Pockels cell and two crossed polarizers can provide exposure times of a few hundred nanoseconds [7]. This approach can be extended to 10 kHz rate using off-the-shelf longitudinal Pockels cell.

In this paper, we introduce the first pulse burst Rayleigh-Raman instrument featuring low-noise, back-illuminated EMCCD cameras operated in subframe burst gating mode [8, 9], to achieve repetition rates of 10 kHz without compromising the measurement precision. The system is demonstrated by conducting simultaneous temperature and methane mole fraction measurements in ambient-temperature gas mixtures and in a non-premixed turbulent inverse diffusion flame. For all test-cases considered, the achieved instrument precision is higher than the previously reported values. The subframe burst gating allows combining kHz rate acquisition to the low-noise performance of CCD cameras. In this mode of operation, only the bottom N pixel rows of the camera chip are exposed and the remaining of the chip is masked. After each laser pulse, the accumulated photocharge is sequentially shifted upward by N rows to the masked region [8, 9]. This process continues until the entire chip is filled. Low-noise, slow-readout through the serial register is then possible, because it does not interfere with the high-speed sampling. A drawback is that the number of recorded images is limited by the camera sensor size. Pulse-burst lasers provide high repetition rate within a burst, but typical burst rates are <0.1 Hz, allowing sufficient time for a slow readout between bursts. Subframe burst gating is therefore an ideal, although unexplored, match for pulse burst laser diagnostics.

Fig. 1 shows the schematic diagram of the optical setup. The different parts of the system are described below. The second harmonic output of a Spectral Energies “Quasimodo” pulse burst laser operated at 10 kHz with 350 mJ/pulse provides the Rayleigh/ Raman excitation source. The maximum burst duration is 10 ms, resulting in a maximum of 100 consecutive pulses. The laser beam is focused at the measurement volume using a 500 mm lens. The

estimated beam spot size is $\sim 100 \mu\text{m}$. Optical breakdown is avoided by operating the laser in “giant pulse” mode, temporally stretching the laser pulse to a full-width at half-maximum of 100 ns. After passing the probe region, the laser beam is re-collimated and directed to a high-speed energy meter (Coherent J-50MT-10KHZ), for shot-to-shot energy correction.

Fig. 1. Schematic diagram of the high-speed Rayleigh-Raman set up.

A pair of air-spaced, three-element achromatic lenses with a diameter of 100 mm, and focal lengths of 400 mm and 300 mm, collects and relays the Rayleigh/Raman signal to the “first image plane”. The lens diameter and focal distance were chosen to fit in the arm of the high-pressure combustion duct at KAUST [10]. The light is then collected by a multi-element Nikon lens of focal length 85 mm and an f-number of 1.8, and collimated to a dichroic mirror with a cut off wavelength at 550 nm. The dichroic mirror splits the collected signal into two: the Rayleigh signal at 532 nm is reflected at 90° to the Rayleigh camera and the Raman signal around 630 nm is transmitted to the Raman camera, as shown in Fig. 1. The cameras are two back-illuminated EMCCD cameras (Princeton Instruments ProEM-HS) synchronized with the laser burst via a delay generator. A multi-element Nikon lens of 50 mm focal length and f-number 1.8 focuses the image on the camera chip. The Raman camera has two filters in front of its collection lens: a 532 nm notch filter of optical density 6 to block any remaining 532 nm light leaking through the dichroic and a bandpass filter with a center wavelength of 635 nm and a bandwidth of 18 nm to pass only the Raman signal of CH stretch region of the CH_4 molecule. A 532 nm bandpass filter with a bandwidth of 10 nm and optical density 4 is placed in front of the collection lens of the Rayleigh camera to cut stray light outside the 532 nm region. Both cameras are placed on a translation stage for fine adjustment of focus. Spatial registration of the two cameras is performed using a pinhole and a target consisting of a dot matrix with $125 \mu\text{m}$ spacing. This step is important since the Raman images at a given spatial location have to be used in conjunction with the Rayleigh images at the corresponding location to obtain accurate and meaningful results. The total magnification of the collection system is ~ 0.79 on both cameras. The entire collection optics is covered with black-out materials to avoid stray light.

For operation in the subframe burst gating mode, generally a mask is applied directly to the chip [8, 9, 11], but this strategy prevents from using the camera in the full frame mode in other experiments. To maintain this capability, in our configuration the masking of the camera is achieved by placing a slit at the first image

plane as illustrated in Fig. 1. This allows higher flexibility because the exposed window can be changed easily by varying the opening of the slit. In our experiment, a window height of 25×1024 pixels is used. With a resolution of 1024×1024 and an equally big frame transfer area, this window size allows a total of 81 images to be acquired during a single burst. The window height of 25 pixels in the vertical direction on the camera, corresponding to a height of $\sim 410 \mu\text{m}$ in the target region, makes sure that the signal is not clipped by the modest beam steering observed during a burst. A major advantage of CCD cameras compared to CMOS cameras is the possibility of hardware binning for superior noise reduction compared to software binning. Hardware binning over the 25-pixel height of the acquisition window is performed to boost the CH_4 Raman signal. The signal on the Rayleigh camera is sufficiently strong and no hardware binning is required. This results in a larger dynamic range, and also allows us to verify that the Rayleigh signal was completely within the slit opening, despite beam-steering and changes in the focusing during the burst. Slow readout (1 MHz) CCD mode provides the best signal-to-noise performance when sufficient light is available, and is used for Rayleigh measurements of temperature, and for measurements of methane mole fraction in non-reacting jets. The Raman camera is operated in the EMCCD mode with a gain of 80 for measurements in the flame, sacrificing instrument precision for lower detection limits.

A non-premixed turbulent inverse diffusion flame is used to demonstrate the instrument in a reactive environment. The burner consists of two concentric stainless steel tubes - the central tube (ID 10.7 mm, OD 12.6 mm) supplies air and the outer tube (ID 15 mm, OD 19 mm) supplies fuel. This is a modified version (larger tube diameters) of the flame investigated in [12]. A tailored mixture of hydrogen, methane and nitrogen in the ratio 33.2 %, 22.1 % and 44.7 % respectively (by volume) is chosen as fuel to provide an approximately constant effective Rayleigh cross section across the flame [13]. The fuel flow rate is 6 standard liters per minute (SLPM) and the central air flow rate is 85.7 SLPM corresponding to a Reynolds number of 12000. A coflow of air at 296 SLPM provides a laminar shroud surrounding the flame. Measurements are conducted at three different heights – 5 mm, 10 mm and 20 mm - from the fuel tube exit.

The signal collected on the Rayleigh camera is software-binned in the vertical and horizontal direction (bin size: 25 and 6, respectively) to improve the SNR and to obtain a probe volume size of $100 \mu\text{m}$. The average of 100 images taken in presence of the flame, and with the laser blocked, is binned and subtracted to each Rayleigh image to account for the camera offset, residual ambient illumination and flame luminosity. The exposure time is $100 \mu\text{s}$, and the corresponding flame luminosity signal is negligible compared to the Rayleigh scattering signal. The Rayleigh signal is then scaled by the instantaneous energy meter reading to account for laser energy fluctuations. Measurements in dry air at a known temperature provide the needed calibration curve Ray_{air} . If T_{air} is the temperature in dry air measured with a thermocouple, Ray is the measured Rayleigh signal, σ_n the normalized Rayleigh cross section defined as the ratio of the Rayleigh cross section of the sample mixture and of air, then the temperature is given by:

$$T = T_{air} \frac{Ray_{air}}{Ray} \sigma_n \quad (1)$$

For the tailored mixture, the Rayleigh cross section across the flame can be approximated to be a constant ($\sigma_n \sim 1$), decoupling

temperature and species measurements. The same binning and background correction is applied to the image collected on the Raman camera to obtain the Raman signal (Ram). A 1D calibration curve Ram_{CH_4} is obtained from measurements in a uniform flow of methane. The temperature in the methane flow T_{CH_4} is measured using a thermocouple. Then the mole fraction of methane is:

$$X_{CH_4} = \frac{T}{T_{CH_4}} \frac{Ram}{Ram_{CH_4}} c(T) \quad (2)$$

where T is the temperature measured from Rayleigh scattering and $c(T)$ is an empirical polynomial that takes into account the change in the integrated intensity of the density-weighted methane Raman spectrum as a function of temperature [14].

The flame luminosity at its peak is 1% of the Raman signal in the unburnt tailored mixture, therefore it cannot be neglected. Since the Raman signal is inversely proportional to temperature, the effect of flame luminosity on the mole fraction is approximately equal to $0.0022 \frac{T}{T_{cold}}$. The flame luminosity is recorded at each location, and subtracted from the Raman signal. Luminosity fluctuations are less than $\pm 10\%$ for the flame considered, leading to a ± 0.0012 error in the single-shot mole fraction, which is much smaller than other sources of error.

Fig. 2. Variation of (a) σ_n vs normalized Raman signal, (b) temperature vs σ_n for various strain rates. The blue curves indicates the region where the normalized Raman signal is less than 2%.

The use of a tailored mixture decouples the temperature measurements from the species measurements, so that the precision of the proposed high-speed Raman and Rayleigh can be assessed independently. Simultaneous Rayleigh/ CH_4 Raman at 10 Hz has been used in the past to obtain temperature measurements in non-premixed jet flames [15]. The sample composition, necessary to compute the Rayleigh cross section, was estimated by combining the fuel number density measurements to a flamelet model. In this work we use the same approach to improve the accuracy of the temperature/species measurements obtained using the tailored mixture. Laminar flame calculations of counterflow diffusion flames for different strain rates are shown in Fig. 2. Fig. 2(a) shows the Rayleigh cross section as a function of the Raman signal normalized by their respective values in the cold tailored mixture. For a given normalized Raman signal, the Rayleigh cross section varies only within $\pm 0.2\%$ for different strain rates, providing an improved estimate of the Rayleigh cross section. When the normalized Raman signal is less than 2%, the Rayleigh cross section is estimated from the temperature, based on Fig. 2(b) (blue region). The average peak temperature at the probed location is used to select the strain rate of the 1D counterflow flame

calculations used for the correction. For the tailored mixture, the effect of this correction is small: ~ -40 K at most in temperature and $\sim .007$ in CH_4 mole fraction, but it would be critical for non-tailored fuels where the Rayleigh cross section doubles going from air to fuel.

Preliminary measurements in ambient air were performed to directly compare the precision of 1D, 10 kHz Rayleigh measurements using the EMCCD camera (Princeton ProEM-HS) operated in the subframe burst gating mode and a high-speed CMOS camera (LaVision Imager pro HS). Energy fluctuations were monitored using a photodiode (Thorlabs DET025A). The energy of the laser was reduced to $1/6^{th}$ of the available laser energy to reproduce the signal level expected in a flame at 1800 K. The signal was software binned to provide a spatial resolution of $78 \mu m$ and $77 \mu m$ on the EMCCD and CMOS cameras respectively. The coefficient of variation (COV), defined as the standard deviation over a single laser burst expressed as the percentage of the mean, is used here as an indication of the instrument precision. For equal conditions, the EMCCD camera achieved a COV of $< 1\%$ that is half of what was achieved with the employed mid-range high-speed CMOS camera (2%). Note that this is not a general result, but highly specific to the experimental set-up used, and to the intensity of the collected signal. CMOS performances approach those of CCD cameras as the signal amplitude increases. As an example Hoffmeister [4] reported a COV of 1% for 10 kHz measurements in air and of $> 2\%$ for measurements in the product region of a laminar flat flame at a temperature of ~ 1100 K due to the lower signal level. By replacing the photodiode with a pyroelectric high-speed energy meter, monitoring of the laser energy fluctuations improved, and a COV of 0.45% was obtained when using the EMCCD camera. This is very close to the 0.4% reported in state-of-the-art 5 Hz Raman/Rayleigh system [14]. To our knowledge, this is the lowest COV reported for Rayleigh measurements in air at 10 kHz.

The Raman signal is significantly weaker than the Rayleigh signal, and measurements with the CMOS camera were too noisy to process even in pure jets of methane at ambient temperature. Previously reported high-speed Raman measurements were taken using high-speed IRO in front of the CMOS camera [4, 5]. It has been extensively reported that the SNR from an ICCD camera is better than for an EMCCD camera only at very low-light conditions (1-10 photons/pixel) [6, 16]. For measurements of CO in flames at 10 Hz, Ajrouche [6] reported a COV of 9.2 % using a back-illuminated CCD camera, 11.2% using a back-illuminated EMCCD, and 17.5% when using an ICCD. For measurements in a uniform flow of methane, we recorded a COV of 0.6% when the EMCCD camera is operated in low-noise CCD mode and 1.2% when operated in EMCCD mode, confirming that the EM-mode does not improve the precision other than for very weak signal. For Raman measurements in flames, notwithstanding the lower precision, the EMCCD mode was preferred to ensure sufficient Raman signal within the flame where the methane concentration is very low. Measurements of methane in a tailored mixture containing 22.1% of methane have a COV of 1.8% using the EMCCD mode with a gain of 80. As a reference, Hoffmeister [4] reported a 3.3% COV for nitrogen in air when using the IRO+CMOS combination, higher laser energies and more efficient collection. Fig. 3 shows the mean and standard deviation of the temperature and mole fraction measurements at three different heights in the flame. The flame is symmetric and the measurements on only one side of the flame are shown. Scatter plots of CH_4 mole fraction versus temperature are also shown in this figure. 10 to 15 burst measurements were taken at each location, depending on the

flow condition to obtain statistically significant measurements of mean and standard deviation.

Fig. 3. (a) The mean and standard deviation of temperature and methane mole fraction, and (b) the scatter plot between methane mole fraction and temperature at different heights for the flame.

At 5 mm height from the burner exit, mixing between the center jet and the fuel annular stream has the effect of partially premixing the fuel mixture, lowering the methane concentration. A laminar flame is formed at the interface between the partially premixed fuel jet and the slow-moving co-flow of air. This is confirmed by the good agreement between the scatter plot with the numerical results of a laminar opposed non-premixed flame with a composition of 16% methane to match the measured peak value of mole fraction. The methane standard deviation profiles peaks at the mixing layer between the turbulent air central jet and the tailored fuel jet (5.5 mm away from the axis). The peak in the temperature standard deviation indicates the mixing between the preheat zone of the laminar flame and the turbulent jet. In correspondence of the peak temperature (>1800 K) the COV is 0.67%, further confirming the higher temperature precision offered by this instrument. As a reference, the Sandia 10 Hz Rayleigh-Raman instrument achieved 0.8% at 2100 K [14], where the pulse-burst 10 kHz system described by Hoffmeister [4] achieved $\sim 2\%$ at 1300 K. The high-precision at elevated temperature further confirms that sub-frame burst gate readout can extend the precision of 10 Hz systems to pulse-burst diagnostics. As we move downstream to 10 mm and 20 mm heights, the flow becomes more turbulent and increased mixing of the fuel with the central air jet reduces the CH_4 mole fraction to <0.05 and the standard deviation in mole fraction increases to $>50\%$ of the mean. The temperature profile is significantly broadened, and the peak temperature becomes lower.

Scatter plot at the downstream conditions shows data points completely filling the region enveloped by the laminar flame calculations, reflecting the large variation in the composition of the partially premixed jet interacting with the co-flow air. Changes in composition on the fuel side, prevent the use of this approach in inverse diffusion flames with non-tailored fuel mixtures.

In summary, for the first time, we combined sub-frame burst gate readout to pulse-burst Rayleigh- CH_4 Raman, to achieve high-speed data acquisition with no loss of precision. The measured temperature precision of 0.45% in air and 0.67% at flame temperature 1800 K are the best precision reported in the literature for 10 kHz Rayleigh measurements. Raman measurements of methane in cold gases had a precision of 0.8% when collected using CCD mode and 1.2 % in EMCCD mode. Measurements have been demonstrated in weakly luminous turbulent flames, and profiles of temperature and methane mole fraction have been presented.

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