High-Speed Linear Raman Spectroscopy for Instability Analysis of a Bluff Body Flame

Jun Kojima
Ohio Aerospace Institute, Brook Park, Ohio

David Fischer
Glenn Research Center, Cleveland, Ohio
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Jun Kojima
Ohio Aerospace Institute, Brook Park, Ohio

David Fischer
Glenn Research Center, Cleveland, Ohio

National Aeronautics and Space Administration

Glenn Research Center
Cleveland, Ohio 44135

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Jun Kojima
Ohio Aerospace Institute
Brook Park, Ohio 44142

David Fischer
National Aeronautics and Space Administration
Glenn Research Center
Cleveland, Ohio 44135

Abstract
We report a high-speed laser diagnostics technique based on point-wise linear Raman spectroscopy for measuring the frequency content of a CH₄-air premixed flame stabilized behind a circular bluff body. The technique, which primarily employs a Nd:YLF pulsed laser and a fast image-intensified CCD camera, successfully measures the time evolution of scalar parameters (N₂, O₂, CH₄, and H₂O) in the vortex-induced flame instability at a data rate of 1 kHz. Oscillation of the V-shaped flame front is quantified through frequency analysis of the combustion species data and their correlations. This technique promises to be a useful diagnostics tool for combustion instability studies.

Introduction
A bluff body flame presents a unique condition for studying flame holding and vortex-flame interactions, which are fundamentally important to many practical combustion devices including industrial boilers and high-speed propulsion (e.g., ramjet) engines (Refs. 1 and 2). The dynamics of vortex-induced instabilities in reacting flows are significantly altered from that in nonreacting flows, known as a von Kármán vortex street (Refs. 3 and 4). This phenomenon is primarily due to a steep density gradient in the thin flame front and sudden gas expansion toward the hot side. The unsteady nature of the phenomenon is often considered as a fundamental subject for computer combustion code validation studies such as large-eddy simulations (Ref. 5). Local flame extinction and blow-off are also common subjects that have brought increasing attention toward bluff body flames (Refs. 1 and 2).

Flow visualization is probably the most popular experimental approach to understanding the complex flow structure behind a bluff body. Various techniques, such as schlieren photography and particle image velocimetry (PIV), have made significant contributions in this area (Refs. 2, 6 to 8). However, since the combustion flow involves rapid chemical reaction and heat transfer along with the vortex-shedding process, time- and space-resolved measurements of combustion species and temperature are essential. Planar laser induced fluorescence (PLIF) imaging of minor species such as OH has served as a powerful tool to visualize flame structure (Ref. 9). Raman spectroscopy is also a highly popular method in this area of diagnostics (Refs. 10 and 11). Nonlinear Raman techniques such as stimulated Raman spectroscopy or coherent anti-Stokes Raman spectroscopy (CARS) (Refs. 12 to 14) provide higher signal intensity and, consequently, better accuracy compared to linear (or spontaneous) Raman techniques, which must work with inherently weaker signals and thus require much higher laser excitation power. On the other hand, linear Raman techniques have a major advantage over nonlinear techniques in that they are capable of simultaneously measuring multiple major species and temperature in the combustion flame with a single laser frequency and with a diagnostic apparatus that is much simpler (Refs. 11, 15 to 17). Also, the fact that spontaneous scattering is linearly proportional to the molecular number density and hence the pressure in combustion systems is useful in practice.
A logical progression in capturing such unsteady combustion behavior is an increase in the temporal sampling rate of the diagnostic system (Refs. 18 to 25). To this end, we have explored the development of a high-speed laser diagnostics system that operates at a sample rate of 1 kHz or greater. A series of important efforts with short-pulse CARS has been reported in this area (Refs. 26 to 28). In general, there is a trade-off in high-speed laser diagnostics between data quality (uncertainty), data rate, dimensions of measurement volume (point-wise versus planar), and number of scalars measured (single or multiple). From our point of view, the data produced by any of these diagnostic methods complement each other when the measurement strategy is well planned and conducted. Our approach, which we describe herein, is to employ a kilohertz-rate, point-wise linear Raman spectroscopy system to simultaneously analyze the temporal behavior of multiple scalars and their interactions. It is critical to measure combustion species and variables indicative of temperature at the same time, since they are directly related to local kinematic viscosity and reaction rate of the fluid in the wake, which are very useful in investigating instability near the shear layer.

**Experimental**

**Bluff Body V-Flame**

Vortex shedding dynamics can be studied in many different configurations with various types of bluff body shapes and dimensions over a wide range of Reynolds number. In this experiment we used a circular cylinder (steel rod), 6 mm in diameter, as a bluff body to stabilize a low-speed flame as shown in Figure 1(a) and (b). The rod was placed approximately 15 mm above a honeycomb flow channel (known as Hencken burner), which generated a homogenized stream of methane gas and air over a 645.2 mm² square area (actual open area 44 percent). The flow rates of the air and the methane fuel were 31 liter/min and 2.9 liter/min, respectively. The Reynolds number was estimated to be $Re = 780$ based on the cylinder diameter and the mean velocity, 2.0 m/s, which was calculated from the total gas flow rate and the open area. Upon ignition, a V-shaped fuel-lean (equivalence ratio $\Phi = 0.89$) CH₄-air premixed flame (Ref. 29) was held to the rod while the von Karman vortex-like wake intrinsically introduced a certain order of oscillation to the thin flame front. Considering the theory of premixed laminar flames, the flame front in our experiment can be seen as a “divider” between the cold unburned mixture and the hot combustion products as illustrated in Figure 1(a). When the flame front fluctuates in space, such spatial movement can be observed through the temporal dynamics at a fixed point, i.e., the probe volume (see the focused laser beam in Figure 1(b)). As seen in Figure 1(b), the V-flame was principally two-dimensional due to a length of cylinder over the honeycomb burner (>25 mm) and the probe volume was placed approximately half way into the full depth of the flame.

**High-Speed Raman Diagnostics**

Figure 2 schematically shows the apparatus for demonstrating high-speed linear Raman spectroscopy in the V-flame. A diode-pumped Nd:YLF laser provided vertically-polarized 527-nm pulses at a repetition rate of 1 kHz (30 mJ/pulse, 210 ns pulse duration, 3 mm beam diameter). The laser beam was focused into the flame with a 700 mm focal length lens. A right-angle folding prism and lens ($f = 400$ mm) redirected the incident beams back into the probe volume, nearly doubling the excitation energy. The Raman scattering signal from the probe volume was collected by a 100 mm focal-length achromatic lens, oriented 90° to the incident beams. Another achromatic lens and retro reflector nearly doubled the collection solid angle of the primary lens, which was mounted back-to-back to a third achromatic lens that imaged the probe volume onto an optical silica fiber assembly for input to a spectrograph. The optical fiber used in this experiment was a bundle of seven 100 µm core (0.22 NA) in a close-pack geometry with 400 µm effective diameter on one end (collection lens side), while the distal end was configured as a 1 by 7 linear array aligned with the slit axis of the spectrograph. The measurement volume was approximately 400 µm long by 400 µm in diameter. The collection fibers
Figure 1.—(a) Bluff body flame (V-flame) experiment configuration. (b) A direct photograph of the V-flame with interrogating Nd:YLF pulsed laser.
Figure 2.—Experimental apparatus showing the high-speed laser Raman diagnostics system, employing a Nd:YLF pulsed laser (527 nm, 1 kHz repetition rate) and a fast image intensified CCD camera (ICCD) coupled with a lens spectrograph. RR: retro-reflector; BD: beam dump.

guided the scattered light to a f/1.8 holographic imaging lens spectrograph equipped with long-wave pass edge filter (535.4 nm) and a volume-transmission grating having 17 nm/mm average reciprocal linear dispersion, and fitted with an image-intensified charge coupled device (ICCD) camera. The camera employed a high-quantum-efficiency Gen III filmless image intensifier (18 mm in diameter) with fast-decay P46 phosphor (<2 µs decay time). The gate width of the image intensifier was set at the laser pulse duration. In order to minimize the readout time and achieve the spectral (frame) rate of 1 kHz, we modified the original 1024 by 256 CCD sensor in software to have an active area of 500 by 30 pixels (each pixel being 26 µm on edge) with 16-bit, 2 MHz digitization while ensuring the necessary spectral range. The spectrograph and the ICCD were optimally assembled to capture a spectrum with a relatively low resolution (ca. 0.38 nm/pixel) over the range from 500 to 690 nm for combustion species detection.

**Results and Discussion**

**Time-Series Scalar Data**

The Raman measurement was made at the probe volume for a period of 30 sec, which yielded 30,000 Raman spectra for a single run. Figure 3 shows the distinct difference between typical single-shot spectra measured on each side of the premixed flame front, i.e., the hot and cold sides. The spectrum of the hot gas is identified by an intense pure-rotational N₂ band around 535 nm and peaks in the vibrational CO₂ and H₂O bands around 580 and 657 nm, respectively. The rotational N₂ band is known to have excellent sensitivity to flame temperature according to our previous work (Ref. 30). In general, the higher the peak of the rotational band in this measurement, the higher the temperature of the gas that is present. The intensity of the rotational N₂ band has, therefore, a semilinear relationship to local temperature. Since CO₂ and H₂O are the major combustion products, it is a clear indicator of the burned gas. In contrast, the spectrum of cold gas has three large peaks from air (oxygen and nitrogen) and fuel CH₄. The vibrational N₂ band at 607 nm in air has a greater peak height due to the vibrational population at room temperature.
Figure 3.—Single-shot spontaneous (linear) Raman scattering spectra in the CH₄-air premixed V flame, showing vibrational bands of the major combustion species and a pure-rotational nitrogen band. Dotted line (blue): cold unburned fuel/air mixture. Solid line (red): hot combustion gas.

The vibrational CH₄ band around 630 nm is visible only in the cold fuel-air mixture as the molecule is quickly broken down in the preheating stage or the earliest stages of the combustion reaction process.

To quantify the oscillating flame front, we first performed super-pixel integrations (i.e., binning) of the above-mentioned Raman bands on a single-shot basis and plotted them as a function of time. The super-pixel region for each band was defined so there was minimum uncertainty from background noise. Figure 4 shows temporal variations of the four Raman bands, each of which represents a molecular species or a scalar, over a period of 700 ms (700 shots). Notice that the scalar signals are interrelated implicitly, as they responded collectively to large or small movements of the flame front. By looking at the peak locations and inter-arrival times, it is easily seen that there is a proportional relationship between the O₂ signal and the CH₄ signal as a result of the co-existence of air and the fuel in unburnt reactants. The CH₄ signal (i.e., concentration) exhibits more frequent (fine) variations than the O₂ signal in several instances of flame front passing. This is seemingly a characteristic behavior of the CH₄ responding to the vortex-flame interaction, in which spatial profiles of the O₂ and CH₄ can be altered from the elementary laminar flame structure (Ref. 31). The profile thickness of the CH₄ is 20 percent thinner than the O₂ based on the maximum gradient, which indicates that the CH₄ may be slightly more sensitive to local flame dynamics and topology. Note that the O₂ signal level does not reach zero even when the CH₄ has zero concentration because the combustion is under a fuel-lean condition and thus has excess oxygen. An apparent inverse motion of the rotational N₂ signal against the O₂ and the CH₄ is also noticeable. This is well understood by the fact that the rotational N₂ signal is a strong temperature indicator (Ref. 30) and that in premixed combustion the fuel consumption rate is directly related to the heat release and thus the resulting flame temperature (Ref. 31). The signal variation of H₂O, a combustion product, supports these ideas by positively following the temperature variations, although the signal has smaller visibility due to the weaker scattering cross-section.
Instability Analysis

We now attempt to quantify the underlying temporal variation (frequency components) of the flame instability behind the circular bluff body. Figure 5 shows the discrete Fourier transforms of the scalar time-domain signals, computed with a fast Fourier transform (FFT) algorithm. Considering the data sampled at 1 kHz, the maximum frequency our system can resolve is limited to \(<500\) Hz. This is well within our region of interest because the Kelvin–Helmholtz instability frequency that is associated with the shear layer of our bluff body flow in this experiment was estimated to be 143 Hz (Ref. 2). This frequency reasonably agrees with the tail end of the amplitude spectrum of each species in Figure 5. According to the amplitude spectra, we found three characteristic frequencies, located at approximately at 5, 27, and 55 Hz, that are almost universal to all of the scalars that we measured. These three frequency components suggest that the wake behind the bluff body caused a series of interactions downstream and generated different modes of instability. The familiar sinuous wake of a bluff body, typically led by enveloped streamlines over the smaller vortices, is probably responsible for the lowest fundamental frequency (~5 Hz) of the present flame instability. It is also reasonable to find that these three modes of frequencies are lower than the estimated highest frequency of von Kármán instability, i.e., 70 Hz because any temperature rise due to combustion reaction should increase the kinematic viscosity of the fluid thus lower the characteristic frequency.

To gain more insight into the instability dynamics, we applied cross-correlation analyses to the time-series data. Figure 6 shows cross-correlations between the CH₄ and the O₂; and H₂O along with an auto-correlation of the CH₄. Firstly, the CH₄ auto-correlation elaborates or adds to the basic findings mentioned above. The time lag (half width of the full maximum) of the fall of the first peak from 1.0 to almost 0.2 is approximately 8 to 10 ms, which represents the width of narrow (intensity) peaks in Figure 4. This
Figure 5.—Fast Fourier Transform (FFT) analysis of the time-series scalar data. Note that there are three fundamental frequencies centered around 5, 27, and 55 Hz.

Figure 6.—Auto- and cross-correlation of temporal fluctuations of the combustion species. Dashed line (blue): auto-correlation of CH₄; Solid line (red): cross-correlation between CH₄ and O₂; Dotted line (green): cross-correlation between CH₄ and H₂O.
timescale is a product of the flame front thickness and the flame propagation speed at the prove volume and detailed discussion would be out of the scope of this paper. The consecutive two peaks in the auto-correlation plot at the time lags of 18 ms, and 36 ms are directly related to the characteristic instability frequencies of 55 Hz (= 1000/18), and 27 Hz (= 1000/36), respectively. Secondly, the cross-correlation of CH₄ and O₂ shows a positive correlation, having almost identical feature with the CH₄ auto-correlation. This is another way of confirming the co-existence and similar behavior of the two gases in unburnt reactants of the premixed flame. Finally, an inverse temporal correlation with little phase shift found in the CH₂H₂O cross-correlation plot is supporting evidence that our bluff body flame is in the so-called flamelet regime (Ref. 32). In the flamelet model, a distinct area separation between the unburnt reactants and combustion products by a thin flame front is assumed for an ease of computational simulation of turbulent flames. Since our instability analysis of the flame front exhibited seesaw-like interrelationships between scalar observables in reactants and that in combustion products, the assumption of thin flame front seems reasonable.

**Conclusion**

In response to the need for advanced experimental techniques that can capture the nature of instability dynamics in combustion, we have developed a high-speed laser Raman spectroscopy system. The diagnostics system consisted of a pulsed Nd:YLF DPSS laser and a customized fast image intensified CCD camera, which achieved a temporal sampling rate of 1 kHz with high signal-to-noise ratio (SNR). We demonstrated the diagnostics technique in a fuel-lean CH₄/air V-flame stabilized behind a circular bluff body (Re = 780), which exhibited a characteristic oscillation at the flame front as a result of vortex-shedding. The temporally- and spatially-resolved measurement process successfully captured time series data of four scalar observables (N₂, O₂, CH₄, and H₂O) in the reacting flow. The temporal variations of the scalar signals indicated oscillating movements of the flame front. We observed a positive correlation between the fuel (CH₄) and oxidizer (O₂) fluctuations, while a negative correlation was found between the reactants and the combustion product (H₂O). We also observed that the thermal fluctuation between the low and high temperatures, indicated by the pure rotational N₂ signal, was correlated strongly with the temporal behavior of the reactants. These experimental observations confirmed that the laminar flame front can be viewed as a thin, high-gradient boundary between the cold unburned mixture and the hot combustion product. This fact, along with the behavior of the cross-correlation functions of the measured scalars, supports the assertion that the flamelet model is a primary combustion regime for the current flame. In addition, we determined the characteristic frequency components of the combustion instability to be 5, 27, and 55 Hz through FFT analysis of the time-series data. This proof-of-concept experiment and the subsequent analysis show that high-speed linear Raman spectroscopy holds promise as a diagnostic tool for combustion instability studies in more practical environments.

**References**

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We report a high-speed laser diagnostics technique based on point-wise linear Raman spectroscopy for measuring the frequency content of a CH4-air premixed flame stabilized behind a circular bluff body. The technique, which primarily employs a Nd:YLF pulsed laser and a fast image-intensified CCD camera, successfully measures the time evolution of scalar parameters (N2, O2, CH4, and H2O) in the vortex-induced flame instability at a data rate of 1 kHz. Oscillation of the V-shaped flame front is quantified through frequency analysis of the combustion species data and their correlations. This technique promises to be a useful diagnostics tool for combustion instability studies.