

## POLARIZED/DEPOLARIZED RAYLEIGH SCATTERING FOR DETERMINING FUEL CONCENTRATIONS IN FLAMES

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Rayleigh scattering has been shown to be a useful diagnostic technique for two-dimensional imaging studies of reacting and non-reacting flows. For example, by combining Rayleigh scattering with a simultaneous measurement of the fuel concentration (e.g., using Raman scattering), mixture fraction and temperature can be determined in flames. In this work, it is demonstrated that the fuel concentration can be obtained by measuring the polarized and depolarized components of the Rayleigh signal and taking their difference or a suitable linear combination. While the depolarized Rayleigh signal is smaller than the polarized signal by a factor of  $\sim 100$ , this is still a factor of  $\sim 10$  larger than the Raman scattering. Application of the technique requires that one of the primary constituents of the fuel stream possess a depolarization ratio sufficiently different from that of the oxidizer. Methane is a convenient candidate as it has no measurable depolarization. Results are shown for methane flames diluted by argon as well as air.

### Introduction

Rayleigh scattering is frequently employed in two-dimensional imaging for temperature and mixture fraction measurements in flames [1–7]. Mixture fraction is defined as the mass fraction of all atoms originating from the fuel stream, and it is a quantity of significant interest to turbulent flame modelers. To calculate mixture fraction, some measure of fuel concentration is required along with the Rayleigh measurement. Species-specific techniques such as Raman scattering or planar laser-induced fluorescence (PLIF) have been used to determine fuel concentration; however, each of these approaches has associated drawbacks. Raman scattering is much weaker than Rayleigh scattering (by  $\sim 10^3$ ) and the modest signal/noise available often requires innovative smoothing approaches [8] and limits the accuracy of the derived mixture fraction. PLIF offers higher signal levels, but requires the use of an additional laser and necessitates tagging the fuel stream with a molecular marker (e.g., acetone). For deriving mixture fraction, the disappearance of the fuel tag well to the rich side of the stoichiometric contour makes this fuel measurement technique unacceptable [3]. This study explores the possibility of measuring the depolarized component of Rayleigh scattering (perpendicular to the laser polarization) in conjunction with the polarized component to infer the fuel concentration in flames. With the larger

cross-section of the depolarized Rayleigh signal (compared to Raman), the combination of these measurements should offer the prospect of improved mixture fraction and temperature imaging in turbulent flames. This paper discusses the basis of the technique and presents calculated and measured signals from laminar flames. Application of the technique to measurements in turbulent flames is discussed in a companion paper [9].

### Polarized and Depolarized Rayleigh Scattering

In most Rayleigh experiments, a linearly polarized laser is used to illuminate the flow, and the Rayleigh-scattered light is collected normal to the illumination line or sheet. Nearly all of the scattered light is polarized parallel to the polarization of the laser with the intensity,  $I_{\parallel}$ , given by the expression

$$I_{\parallel} = KI_0N \sum_i \sigma_i x_i \quad (1)$$

$K$  is the calibration constant of the collection optics,  $I_0$  is the intensity of incident laser light, and  $N$  is the total number of molecules contained in the probe volume. The summation is over all species with  $x_i$  the mole fraction and  $\sigma_i$  the Rayleigh cross-section of the  $i$ th gas in the mixture.

TABLE 1  
Depolarization ratios for some common gases at various wavelengths

Gas	$\rho_p \times 10^2$				
	444 nm	488 nm	514.5 nm	632.8 nm	This work, 532 nm
N <sub>2</sub>	1.3 <sup>a</sup>	1.08 <sup>b</sup>	0.96 ± 0.14 <sup>d</sup>	1.042 ± 0.006 <sup>c</sup>	1.02 ± 0.02
O <sub>2</sub>	3.6 <sup>a</sup>	2.91 <sup>b</sup>	—	3.02 ± 0.01 <sup>e</sup>	2.70 ± 0.10
CO <sub>2</sub>	—	4.12 ± 0.02 <sup>c</sup>	4.085 ± 0.02 <sup>c</sup>	4.05 ± 0.02 <sup>c</sup>	3.91 ± 0.04
CO	—	0.521 ± 0.007 <sup>c</sup>	0.519 ± 0.007 <sup>c</sup>	0.48 ± 0.005 <sup>e</sup>	0.538 ± 0.015
H <sub>2</sub>	—	0.95 <sup>b</sup>	—	0.80 <sup>e</sup>	—
H <sub>2</sub> O	—	—	0.0299 ± 0.0135 <sup>d</sup>	—	—
C <sub>2</sub> H <sub>2</sub>	—	1.897 ± 0.005 <sup>c</sup>	1.889 ± 0.005 <sup>c</sup>	1.851 ± 0.004 <sup>e</sup>	—
C <sub>2</sub> H <sub>4</sub>	—	1.266 ± 0.005 <sup>c</sup>	1.247 ± 0.005 <sup>c</sup>	1.207 ± 0.002 <sup>c</sup>	—
C <sub>2</sub> H <sub>6</sub>	—	0.190 ± 0.003 <sup>c</sup>	0.188 ± 0.004 <sup>c</sup>	0.166 ± 0.001 <sup>c</sup>	—
C <sub>3</sub> H <sub>8</sub>	—	0.214 ± 0.003 <sup>c</sup>	0.208 ± 0.006 <sup>c</sup>	0.195 ± 0.007 <sup>c</sup>	0.181 ± 0.015

<sup>a</sup> Ref. [13].

<sup>b</sup> Ref. [14].

<sup>c</sup> Ref. [15].

<sup>d</sup> Ref. [16].

<sup>f</sup> Ref. [17].

The classical treatment of Rayleigh scattering predicts that a small fraction of polarized incident radiation will become depolarized for scattering objects that are not isotropic (i.e., spherically symmetric) [10]. The depolarization ratio,  $\rho_p$ , is defined as the ratio of the radiant intensities scattered perpendicular ( $I_{\perp}$ ) and parallel ( $I_{\parallel}$ ) to the incoming polarized source (denoted by the subscript  $p$ ), and can be written

$$\rho_p = \frac{I_{\perp}}{I_{\parallel}} = \frac{3\gamma^2}{45\alpha^2 + 4\gamma^2} \quad (2)$$

where  $\alpha$  is the mean value of the polarizability and  $\gamma$  is the anisotropy [11]. All components of the polarizability tensor are frequency dependent, and therefore, the depolarization ratio will vary with the excitation wavelength. Equation 2 represents the depolarization ratio at the exact frequency of the laser; in practice, the measured depolarized signal will also include contributions from the pure rotational Raman spectrum as well (unless high spectral resolution is used) [12]. Table 1 shows literature values of  $\rho_p$  (the total line including rotational Raman lines) for a number of common flame molecules at different wavelengths [13–17]. It is seen from Table 1 that depolarization ratios for most of the major species in typical flames are on the order of a few percent. Notable exceptions are some common fuels, which have significantly smaller depolarization ratios (or for the case of methane, essentially zero depolarization because of the spherical-top molecular configuration). It is the significantly different depolarization ratio of the fuel that will be exploited in obtaining the fuel concentration from the difference between

the polarized and depolarized Rayleigh signals. Using the depolarized Rayleigh signal for concentration measurements has previously been demonstrated in plasmas of simple composition [18].

If values of depolarization ratios, Rayleigh cross-sections, and species concentrations are known, it is possible to predict the signal strength of both the polarized and the depolarized Rayleigh scattering in flames. Counterflow laminar flame calculations are frequently used to predict species concentrations and the results of such calculations can be coupled with known values of  $\rho_p$  and  $\sigma_i$  to investigate the details of the expected signals. As an initial test of the idea of obtaining the fuel concentration from the difference between the polarized and depolarized Rayleigh signals, the case of a fuel mixture of methane and argon is considered. Both methane and argon have zero depolarization and thus represent an ideal case for the technique. The predicted polarized and depolarized Rayleigh signals (plotted as a function of mixture fraction) for a 35% CH<sub>4</sub>/65% Ar (by volume) flame are shown in Fig. 1a. While the polarized Rayleigh signal is maximum at unity mixture fraction, the depolarized signal goes to zero.

The difference ( $I_D$ ) between the parallel and perpendicular polarization of the Rayleigh signal (normalized to values in air) is given by

$$I_D = \frac{I_{\parallel}}{I_{\parallel,\text{air}}} - \frac{I_{\perp}}{I_{\perp,\text{air}}} = \frac{N}{N_0} \left[ \frac{\sum_i \sigma_i x_i}{I_{\parallel,\text{air}}} - \frac{\sum_i \sigma_i \rho_{p,i} x_i}{I_{\perp,\text{air}}} \right] \quad (3)$$

where  $\sigma_i$  is the Rayleigh cross-section,  $x_i$  is the mole

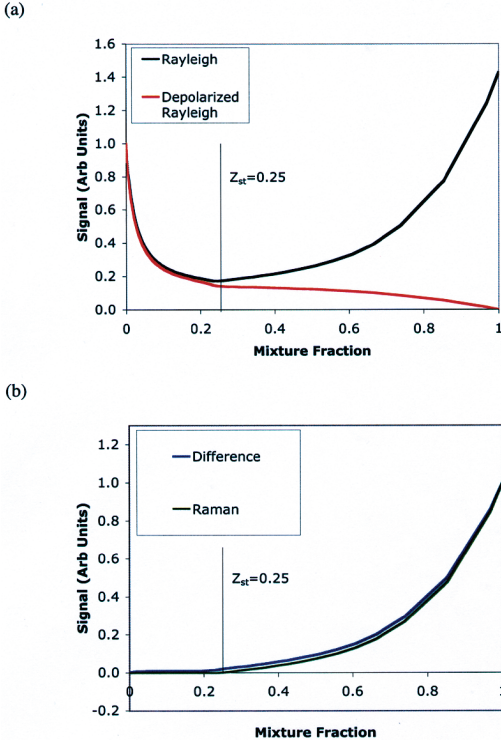


FIG. 1. (a) Computed signal profiles for the polarized and depolarized components of the Rayleigh scattering as a function of mixture fraction in an argon-diluted methane laminar flame. Signals are scaled to have the same value in air (mixture fraction = 0). (b) Computed signal profiles for the methane Raman scattering and the difference Rayleigh scattering as a function of mixture fraction.

fraction of species  $i$ ,  $N$  is the number density at a given point in the flame (inversely proportional to temperature), and  $N_0$  is the number density at the calibration temperature and pressure. Predicted difference Rayleigh and fuel concentration profiles shown in Fig. 1b indicate that  $I_D$  should provide an excellent fuel tracer for this case (with  $\sim 10$  times the signal of the methane Raman in the fuel stream). The calculations include the effect of the changing overall depolarization  $\rho_{p,mix}$  due to composition changes arising from reaction. One primary combustion product is  $\text{CO}_2$ , which has the largest  $\rho_p$  appearing in Table 1. This is balanced by smaller depolarization ratios for  $\text{H}_2\text{O}$  and other products or intermediates appearing in significant concentrations such as  $\text{CO}$ .

In practice, several factors must be quantified in correcting the experimentally obtained signal. The first of these is the nonideality of the polarizing filter

used and the accuracy of its orientation relative to the laser polarization. In the perpendicular imaging mode, some of the parallel polarization signal will pass through the filter element. This effect will be compounded by any depolarization present in the laser source itself and from background scattering. Thus, the measured signal,  $S_{\perp}$ , is a combination of the true depolarization component,  $I_{\perp}$ , background scattering of the same polarization,  $B_{\perp}$  (which must be accounted for separately), and some contribution from  $I_{\parallel}$  arising from the aforementioned factors:

$$S_{\perp} = I_{\perp} + B_{\perp} + (\ell I_{\parallel}) \quad (4)$$

where  $\ell$  accounts for the parallel polarized Rayleigh “leakage” signal or cross-talk. If the background scattering, the response of the imaging system, and the leakage of the polarizer are the same for the measurements of the polarized and depolarized components, and if the contribution of leakage from the (small) depolarized to the (large) polarized component is negligible, the leakage can be expressed as

$$\ell = \frac{S_{\perp,NG1} - S_{\perp,NG2}}{S_{\parallel,NG1} - S_{\parallel,NG2}} \quad (5)$$

where  $S$  is the uncorrected, pixelwise intensity as measured for two gases NG1 and NG2 with zero depolarization, such as noble gases. Thus, from imaging the scattering components of two such gases at known uniform temperature and pressure, it is possible to account for the leakage of the polarized Rayleigh and background scattering into the image of the depolarized scattering. If the background, response, fixed pattern, or polarizer is different for the two images (as is normally the case for simultaneous imaging of the two components), the actual computation becomes somewhat more complicated but follows the same principle. The same calibration images, together with the knowledge of  $\sigma_{NG1}$  and  $\sigma_{NG2}$ , also allow for the usual correction of background scattering of the same polarization in each image.

Data on the depolarization ratios of the major species are not available from literature at 532 nm (second harmonic of Nd:YAG), which was used in the validation experiments described below. Therefore, measurements of these depolarization ratios were performed. The second harmonic of a Nd:YAG laser was focused to a line by a spherical lens with 750 mm focal length. The beam entered and left a metal sample cylinder (50 mm diameter) through small holes. The gas being measured was fed continuously to the cylinder at ambient temperature and pressure. The absence of any laser windows and the use of appropriate irises provided a field of view that was practically free of stray light. The Rayleigh scattering was imaged by a cooled CCD camera (Photometrics,  $512 \times 512$  pixels) with the polarizer (B + W photographic circular polarizer) mounted in front of the objective (Nikor 50 mm,  $f/1.4$ ). The leakage of the

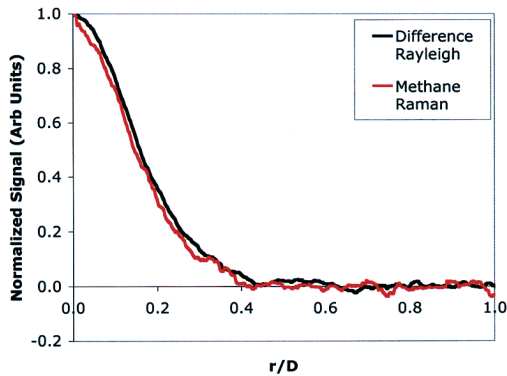


FIG. 2. Experimental measurements of the methane Raman and difference Rayleigh signals as a function of non-dimensional radius ( $D = 16$  mm).

polarizer was determined by taking images of scattering in argon and helium as outlined above, and the depolarized scattering was corrected accordingly, although the leakage of this specific polarizer is negligible in the context of noisier sheet measurements in actual combustion (see below). For the polarized and depolarized components, 50 and 100 shots, respectively, were averaged. The resulting depolarization ratios are given in Table 1. The uncertainty in the depolarization ratios measured has been assessed by estimating uncertainty in the background and leakage corrections and evaluating the variations within the image. The underlying cause of correction uncertainties is the drift in laser power over the time of the measurement series. The measurements were taken such that first the polarized scattering was recorded for all gases, next the polarizer was rotated, and then the depolarized scattering was recorded for all gases. Any error in polarizer alignment is therefore constant within each series and is accounted for by the leakage correction. The scattering from air was measured at the beginning and end of each series to estimate the power drift, which was found to be about 1%. The value for oxygen is calculated from the measurements for nitrogen and air. Values for water vapor and hydrogen were not measured and are taken to be the values available from the literature that are closest to 532 nm.

### Measurements in an Argon-Diluted Methane Flame

The technique was investigated in two different axisymmetric laminar diffusion flames using two different experimental configurations. In an initial set of experiments, images of an argon-diluted methane flame (35%  $\text{CH}_4/65\%$  Ar, jet diameter  $D = 16$  mm) were taken such that both regions of the undiffused

fuel stream and the air of the coflow were captured. The second harmonic of a Nd:YAG laser (Continuum, Powerlite 8000; 260 mJ/pulse) was focused into a sheet by a cylindrical lens (250 mm focal length). Scattered light was collected normal to the laser sheet using a large format  $f/1.4$  camera objective (Nikor 85 mm) and focused onto an unintensified interline transfer CCD camera (PCO/Cooke, SensiCam,  $1024 \times 1280$  pixels). Flame luminosity was suppressed by gating the camera electronically for  $2 \mu\text{s}$ . To further suppress luminosity, an interference filter (10 nm full width at half-maximum [FWHM] centered at 532 nm) was placed between objective and camera, together with a sheet polarizer (Melles Griot, 003FPG), whose orientation was rotated  $90^\circ$  as required. Integration over 12 and 336 shots was performed for the polarized and depolarized components, respectively. Sufficient suppression of background scattering was achieved by directing the laser beam on either side of the flame through tubes with staged baffles. As an independent, direct measurement of the fuel concentration, Raman scattering of methane was imaged successively on the same camera, exchanging polarizer and filter for the appropriate interference filter (10 nm FWHM centered at 630 nm), integrating  $\sim 1000$  shots, and then averaging three images.

Figure 2 shows the measured difference Rayleigh and methane Raman signals as a function of radial position in the 35%  $\text{CH}_4/65\%$  Ar flame. As predicted by the calculations, the two measured signals are essentially identical. The leakage,  $\ell$ , was determined to be 0.82%. The value was determined from uniform-field images of the scattering from argon and helium and was taken as the mean of the pixel-wise leakage calculated from the calibration images. This yields a parasitic contribution to the measured depolarized signal that is on the same order as the expected true depolarized Rayleigh scattering. The corrections remove this contribution reasonably well as verified with images of argon, which should give zero depolarized scattering.

Variations of  $\rho_p$  with temperature can be estimated by evaluating the dependence of the molecular polarizability and anisotropy in equation 2. For diatomic molecules, the mean polarizability ( $\alpha$ ) increases by only 1% for every 1000 K increase in temperature [19,20]. The temperature dependence of the polarizability anisotropy ( $\gamma$ ) has not been established experimentally; however, it is possible to use semitheoretical arguments to assess the appropriate trend. *Ab initio* calculations of the polarizability of  $\text{H}_2$  reveal a larger relative increase in the anisotropy than the polarizability with increasing internuclear distance [21]. The increase is estimated to be on the order of 2% per 1000 K temperature increase. Overall, the temperature dependence of the depolarization ratio will be approximately 2% per 1000 K or no more than 4% for typical flame

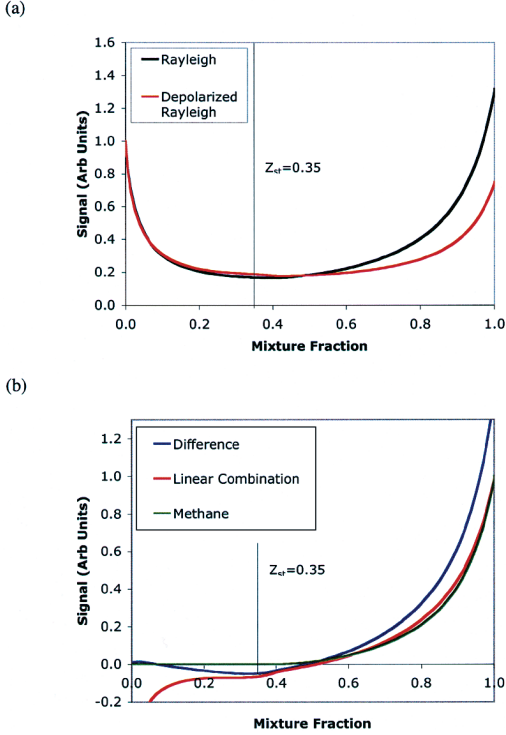


FIG. 3. (a) Computed signal profiles for the polarized and depolarized components of the Rayleigh scattering as a function of mixture fraction in an air-diluted methane laminar flame. Signals are scaled to have the same value in air (mixture fraction = 0). (b) Computed signal profiles for the methane Raman scattering, the difference Rayleigh scattering (which is multivalued) and a linear combination of Rayleigh and depolarized Rayleigh, as a function of mixture fraction.

temperatures of approximately 2000 K. We therefore conclude that temperature effects pose no serious difficulty for the present purpose as long as the difference Rayleigh signal exhibits consistent and predictable behavior through the flame front region.

### Calculations and Measurements in an Air-Diluted Methane Flame

For a particular choice of fuel diluent, the detailed behavior of the difference Rayleigh signal must be considered to infer accurately the fuel concentration (i.e., laminar flame calculations should be performed and the signals modeled as described above). One fuel/diluent mixture of particular interest is the partially premixed methane/air ratio of 25% methane/75% air. In a recent series of workshops on turbulent non-premixed flames [22], considerable work has been done both experimentally and computationally

on flames with this composition. By combining polarized and depolarized measurements in these flames, it should be possible to image the mixture fraction (as well as temperature) and hence determine two components of the scalar dissipation, which has not been done in these flames.

Figure 3a shows the calculated Rayleigh and depolarized Rayleigh signals as a function of mixture fraction for a fuel mixture of 75% air and 25% methane. One notable feature of the calculated signals for this fuel is that in the mixture fraction range from  $\sim 0.2$  to  $0.4$ , the (normalized) depolarized signal is greater than the polarized signal. This leads to a negative region in the difference between the two signals as seen in Fig. 3b. The negative value of the difference signal could be handled by simply adding an offset, but a more significant problem stems from the fact that the difference signal is multivalued with respect to the methane concentration (also shown in Fig. 3b). This would make it impossible to assign the difference signal to a unique value of methane concentration. This problem can be addressed by forming a difference signal based on a different linear combination of the polarized and depolarized signals. In particular, the signal can be written as

$$I_{LC} = \frac{I_{\parallel}}{I_{\parallel,air}} - a \frac{I_{\perp}}{I_{\perp,air}} + b \quad (6)$$

While the simple difference (i.e.,  $a = 1$  and  $b = 0$ ) worked well for the argon-diluted flame, a value of  $a = 1.3$  was needed for the air-diluted flame to provide a single-valued function with respect to the methane signal. By choosing an offset of  $b = 0.065$ , the resulting linear combination signal becomes negative at nearly the same mixture fraction value that the methane concentration goes to zero as seen in Fig. 3b. In using the signal then, a unique mapping of the linear combination signal onto the fuel concentration can be performed.

A second set of experiments was performed to check the applicability of the technique for this fuel composition. The measurements were performed on the air-diluted “flame A” of the “International Workshop on Measurement and Computation of Turbulent Nonpremixed Flames” [22] (25%  $\text{CH}_4$ /75% air, jet diameter  $D = 7.2$  mm). The beams of two frequency-doubled Nd:YAG lasers (560 and 360 mJ/Pulse) were combined geometrically and retro-reflected to maximize power. The polarized component was imaged by the same electronically gated camera as above, with the objective (Nikor 50 mm,  $f/1.4$ ) mounted directly to the camera. The interference filter was placed in front of the objective. Since this imaging train was solely devoted to measuring the (predominant) polarized component, no polarizer was used, potentially resulting in an error on the order of 1% due to the depolarized component, but increasing the total transmission to the camera significantly. Imaging of the depolarized component

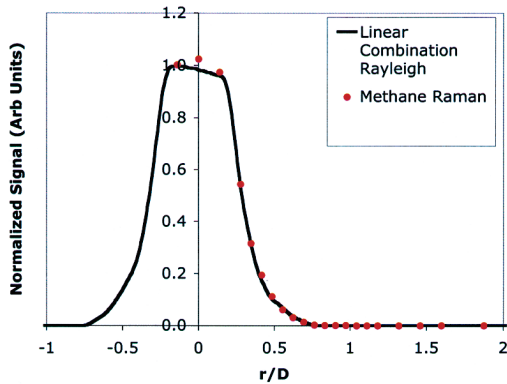


FIG. 4. Experimental measurements of the methane Raman scattering and a linear combination of the polarized and depolarized Rayleigh signals as a function of non-dimensional radius ( $D = 7.2$  mm). In the latter, negative values are set to zero.

was achieved by optically coupling a similar interline transfer CCD camera (PCO/Cooke, SensiCam,  $320 \times 240$  pixels after  $2 \times 2$  binning) to an image intensifier (ITT Gen III). An interference filter in front of the objective (85 mm  $f/1.2$ ) and the short gating time of the intensifier (400 ns) suppressed luminosity. Here, a different polarizer (B + W photographic circular polarizer) was placed in front of the objective and could be rotated to be able to take the necessary calibration images. Synchronization and registration of the system are described in Ref. [9]. For each image, 80 shots were averaged.

In the second experiment, the leakage was much smaller ( $\ell = 0.04\%$ ), due to the use of the different polarizer. Therefore, the parasitic contribution is reduced to about 4% of the expected true depolarized Rayleigh scattering and was neglected. Omitting the corrections for cross-talk yields significant simplification of the data analysis for this two-camera configuration.

Figure 4 shows the measured linear combination of the polarized and depolarized Rayleigh signals as a function of radius in flame A. Also shown in the figure is the measured fuel concentration obtained previously by single-point Raman scattering [23]. For the comparison, the Rayleigh signals have been smoothed to match the spatial resolution of the Raman measurements ( $0.5 \times 0.5 \times 0.5$  mm<sup>3</sup>). Once again the agreement is extremely good, demonstrating the applicability of the technique even for relatively dilute fuel mixtures.

Even with an order of magnitude increase in cross-section of the depolarized Rayleigh over the corresponding Raman scattering, the resultant signals are still relatively weak. For the experimental configuration used for measurements in the air-diluted

methane flame, the signal-to-noise ratio of the depolarized Rayleigh in single-shot images of ambient air was  $\sim 10$ . This necessitated some degree of smoothing to determine mixture fraction reliably from single-shot images. The application of this technique to turbulent flames is described in Ref. [9]. In those experiments, the polarized/depolarized Rayleigh measurements were combined with simultaneous imaging of OH and CO, which further helped to quantify the mixture fraction near the flame front. However, if the experiment were optimized for polarized/depolarized measurement alone (e.g., by using an intracavity configuration providing greater laser-sheet intensity), it should be possible to further improve the signal-to-noise ratio.

## Conclusions

The present work establishes depolarization Rayleigh scattering as a viable diagnostic technique for measurements of fuel concentration in both argon-diluted (65% by volume) and air-diluted (75% by volume) laminar methane flames. The difference Rayleigh signal increases the postprocessed signal-to-noise ratio of the effective fuel image by more than a factor of 3 along the jet centerline. This technique can be applied in situations where one or more primary molecular constituents (in this case, methane) possess a significant deviation in depolarization ratio from that of the oxidizer coflow. Table 1 indicates that other candidate fuels might include ethane and propane, which both have small depolarization ratios relative to air. Dilution of the fuel with gases such as argon or helium will allow extension of this technique to higher  $\rho_p$  fuel molecules. Furthermore, such dilution is favorable in that it will increase the dynamic range of the difference signal between the fuel mixture and air coflow. The presence of broadband fluorescence interferences (which are unpolarized and would significantly influence the depolarized Rayleigh signal) arising from polycyclic aromatic hydrocarbons may pose a problem in some flames and should be examined prior to application.

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## COMMENTS

*Katharina Kohse-Hoeinghaus, Bielefeld University, Germany.* Your technique offers a better signal-to-noise ratio than Raman measurements, which would, however, still be possible in a methane flame and would have the advantage of being direct rather than indirect. Could you comment on the potential of using this approach in flames of, for example, higher hydrocarbon fuels where Raman measurements would not be feasible?

*Author's Reply.* We have not done the experiments or laminar flame calculations for fuels other than methane. However, we believe the technique may indeed work for other fuels. Table 1 shows that the depolarization ratios of ethane and propane, while not zero, are considerably lower than that of air, making those good candidates. In addition, if the fuel from the jet contains a significant fraction of a noble gas, then the overall depolarization ratio of the mixture will also be small compared to that of air and products, resulting in a difference signal that is representative of the fuel concentration.

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*Yung-Cheng Chen, The University of Sydney, Australia.* It was stated in the presentation that the CCD camera for polarized Rayleigh scattering does not use an image intensifier, for better spatial resolution. However, the camera for depolarized Rayleigh scattering is intensified. Will the final image spatial resolution for the mixture fraction improve by using a non-intensified CCD camera for polarized Rayleigh scattering?

*Author's Reply.* Yes. The polarized image, in addition to giving information on the temperature, is used in smoothing the depolarized image (Ref. [8] in paper). Therefore, improving the spatial resolution in the polarized image by using an unintensified camera helps improve the resolution of both the temperature and the fuel concentration fields, which are needed to derive the mixture fraction.