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Background-Free Species Detection in Sooty Flames Using Degenerate Four-Wave Mixing

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The background radiation disturbance in luminous environments has been compared for degenerate four-wave mixing (DFWM) and laser-induced fluorescence (LIF) for OH radical detection in a sooty propane/oxygen flame. The LIF signal generally was considerably stronger than the DFWM signal, but in strongly sooty environments the LIF signal was accompanied by a significant background signal, while the DFWM signal was background-free under all soot loads tested.

Index Headings: Combustion diagnostics; Degenerate four-wave mixing; Laser-induced fluorescence.

INTRODUCTION

The techniques predominantly used in laser diagnostics in combustion are laser-induced fluorescence (LIF) and CARS.¹ For temperature and species concentration determination, these methods can often provide the information required. Nevertheless, there are still a variety of species and measurement conditions for which information is difficult to retrieve with these techniques as well as with other established methods (absorption spectroscopy, Raman or Rayleigh scattering, etc.). There is therefore a continuous development and application of new methods for laser diagnostics within the combustion community. A few years ago^{2,3} it was realized that degenerate four-wave mixing (DFWM) could be useful in combustion diagnostics, and it has since been developed for this purpose (e.g., at Oxford^{4,5} and Sandia National Laboratories^{6,7}). Considering the fact that all four photons in a DFWM process induce transitions between states connected with allowed electric dipole transitions (for the moment neglecting two-photon DFWM⁸), its detection sensitivity should be similar to that of doubly resonant CARS. As in CARS, the signal is also emitted as a collimated coherent laser beam. However, this approach would generally compete favorably with doubly resonant CARS for species detection due to its relative simplicity, as only one excitation wavelength is used. As DFWM primarily is a method for species concentration determination (although it has recently also been used for temperature determination^{9,10}), it mainly competes with LIF. As DFWM is intrinsically a multi-photon technique, it would generally not be quite as sensitive as LIF. It could, however, compete favorably if, for example, the measurement region had a geometry preventing fluorescence detection or only allowing fluorescence detection with low efficiency, such as when the measurement point is situated far away from the detection equipment. Another situation where DFWM potentially could be a re-

alistic alternative to LIF is when there is a strong optical background, spectrally and temporally coincident with the fluorescence signal. As the information in DFWM propagates as a collimated beam, background radiation can be eliminated also by spatial filtering. This last situation applies to sooty flames, and this paper compares detection of OH in a sooty propane/oxygen flame using LIF and DFWM.

EXPERIMENTAL SETUP AND MEASUREMENTS

The burner was a welding torch with a 1.5-mm nozzle diameter, and a premixed C₃H₈/O₂ flame was used. The propane flow velocity at the burner nozzle was 2.7 m/s, and the oxygen flow velocity ranged from 0 to 3.7 m/s. On the basis of the flow rates, the C/O ratio in the fuel oxygen mixture ranged from infinity down to 1.1 at the highest oxygen flow. That is, all oxygen flows used in this measurement corresponded to C/O values well above the soot point, which is approximately 0.5 for propane. However, for such high C/O ratios the burner simultaneously functions as a diffusion burner, and the C/O ratios calculated from the gas flow therefore have little or no physical significance.

The optical layout is shown in Fig. 1. The image magnification for the LIF detection was a factor of two and the collection angle was approximately 0.05 sr. The laser system consisted of a Quantel YG581-10 TEM₀₀ YAG laser pumping a Quantel TDL50 dye laser. The excitation pulse energy was (typically) 2 mJ in a 5-mm-diameter beam. The dye laser linewidth was approximately 0.7 cm⁻¹. Four percent of the pulse energy was split off for the probe beam, which was crossed with the pump beam at an angle of 7°. The pump beam was retroreflected back through the flame, retracing its own path, as shown in Fig. 1. Twenty percent of the DFWM signal was directed through a spatial filter to an EG&G PARC diode array detector, which was also used for detecting the LIF signal. The DFWM and LIF signals were imaged onto different parts of the diode array and could therefore be detected simultaneously. However, during data recording one of the signals was always blocked, and during the LIF measurements also the probe beam and the retroreflected pump beam were blocked. Although not explicitly shown in Fig. 1, the complete interaction region between the laser beams and the flame was imaged on the diode array.

The OH R₁(8) and R₁(10) transitions in the A²Σ-X²Π v'' = 0 → v' = 0 band at λ = 306.3 nm and the R₁(9) transition in the A²Σ-X²Π v'' = 0 → v' = 1 band at λ = 282 nm were investigated. In both cases detection was performed with the use of a filter with 27% transmission at λ = 308.9 nm and with an FWHM of 11.5 nm. For the

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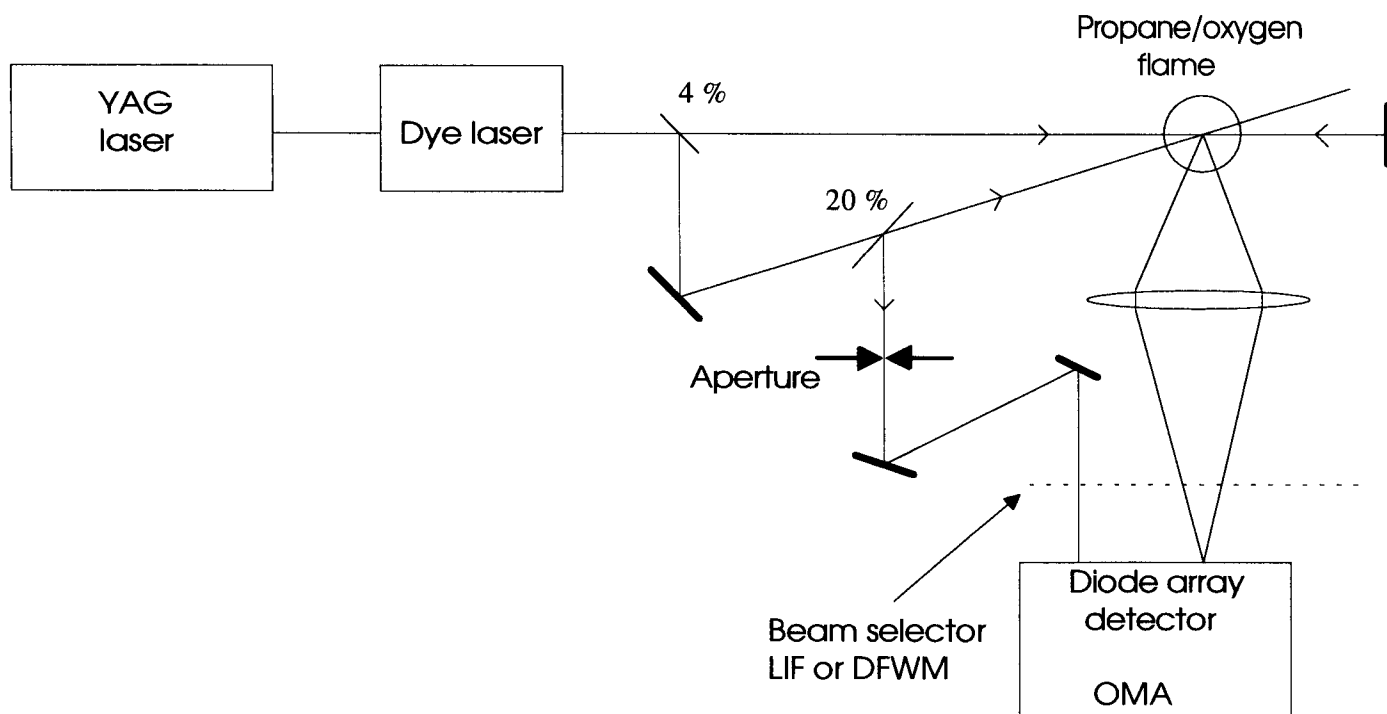


FIG. 1. Experimental setup for simultaneous LIF and DFWM detection.

$v'' = 0 \rightarrow v' = 1$ transition, detection at 330 nm was briefly tested and found to be inferior to 309-nm detection. For the 0-0 band LIF and DFWM measurements were performed both 1 cm and 5 cm above the burner nozzle. For the 0-1 transition only LIF measurements were performed, and that at a distance of 5 cm above the nozzle. At the beginning and end of every measurement series LIF and DFWM signals were recorded with the flame turned off.

RESULTS AND DISCUSSION

DFWM and LIF signals for excitation on the 0-0 band 1 cm above the burner nozzle are shown vs. oxygen flow in Fig. 2. For each oxygen flow setting both LIF and DFWM recordings were performed before the flow was changed. Three such runs are shown, and the spread between the points illustrates the statistical scatter of data. In these measurements neither LIF nor DFWM background was seen. Figure 3 shows the DFWM signal, the LIF signal, and the LIF background 5 cm above the burner. The LIF background was obtained by tuning the laser off the 0-0 band resonance. With the flame turned off there was no detectable background on the diode array either with the use of LIF or with the use of DFWM. Each measurement series was then recorded with the flame burning and the laser first tuned on resonance and then off resonance. With the laser tuned off resonance there was no DFWM background. There was no LIF background 1 cm above the burner nozzle but significant LIF background 5 cm above the nozzle both for 0-0 and 0-1 band excitation. To utilize the strength of LIF, where exciting laser radiation and detected fluorescence do not necessarily have to be on the same wavelength, we also investigated the LIF signal and background for $v'' =$

$0 \rightarrow v' = 1$ band excitation and $v' = 0 \rightarrow v'' = 0$ band detection. Here, the LIF signal was a factor of ten lower since the line strength of the 0-1 band transition is weaker than the 0-0 band line strength, but the background, although still significant, had decreased to less than 40% of the LIF signal at the lowest oxygen flow and to only a few percent at the higher flows.

As can be seen from Figs. 2 and 3, DFWM generally has lower signal strength than LIF even at comparatively high OH densities. (The OH radical density is estimated to be approximately $10^{16}/\text{cm}^3$; see, for example, Ref. 11.) However, DFWM can be essentially background-free in luminous environments where LIF may be embedded in significant background radiation, as illustrated in Fig. 3.

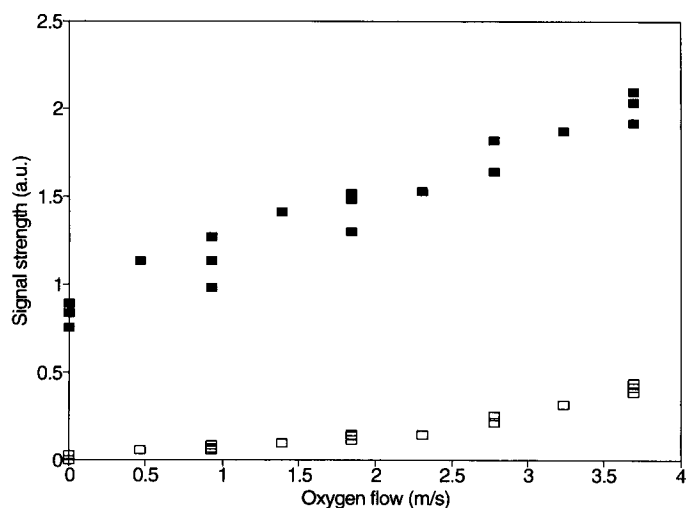


FIG. 2. DFWM (open symbols) and LIF signals (filled symbols) 1 cm above burner nozzle.

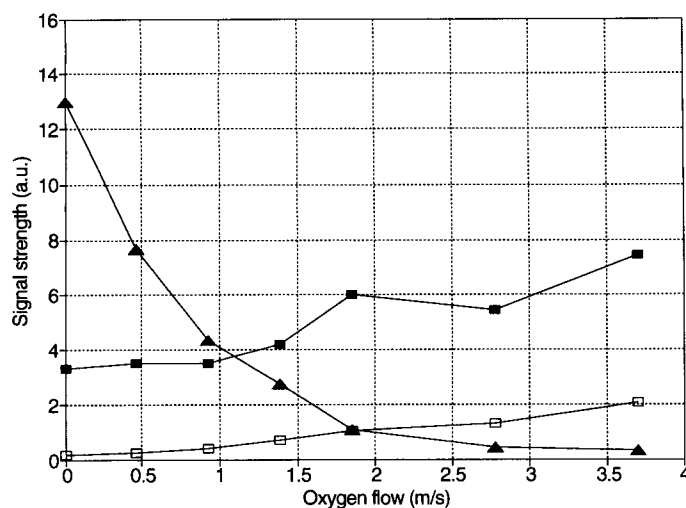


FIG. 3. DFWM (open squares) and LIF signals (filled squares) 5 cm above burner nozzle. The LIF measurements were perturbed by significant background from scattered laser light (triangles).

Since the DFWM signal is emitted as a collimated laser beam, background radiation can be efficiently eliminated by spatial filtering also when it is temporally and spectrally coincident with the signal. To some extent, spatial discrimination can also be performed in LIF measurements; however, in turbulent flames this is not generally possible. We believe that the LIF background signal in Fig. 3, where detection is made at the laser wavelength, mainly originates from elastic scattering of soot particles. Another possibility would be rotational Raman scattering.¹¹ One centimeter above the burner surface, as in Fig. 2, the particles are still too small to scatter so much light,

so that it is comparable to the LIF signal. For 0–1 band excitation at 282 nm where the 0–0 band detection at 307 nm is on the red side of the exciting laser light, it instead seems reasonable to assume that the background originates from polyaromatic hydrocarbons.¹² Existence of PAHs 5 cm above the burner nozzle could in principle also result in a DFWM background. This outcome was, however, not observed, indicating the DFWM is not very sensitive to such disturbances.

This investigation has illustrated the immunity of DFWM, as compared to LIF, to background radiation disturbance in luminous environments. It may be concluded that DFWM is a viable alternative to LIF in these situations, although not necessarily better.

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