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A00-16621

AIAA 2000-0777

Quantitative, High Sensitivity Diagnostic of Combustion Radicals using Wavelength-Modulated UV Sources

K. A. Peterson and D. B. Oh
Southwest Sciences, Inc.
1570 Pacheco St., Suite E-11
Santa Fe, NM 87505

38th Aerospace Sciences Meeting & Exhibit
10-13 January 2000 / Reno, NV

QUANTITATIVE, HIGH SENSITIVITY DIAGNOSTICS OF COMBUSTION RADICALS USING WAVELENGTH-MODULATED UV SOURCES

K. A. Peterson[*] and D. B. Oh[†]
Southwest Sciences, Inc.
1570 Pacheco St., Suite E-11
Santa Fe, NM 87505

ABSTRACT

The application of wavelength modulated, tunable UV sources to combustion radical diagnostics is described. Tunable, wavelength modulated UV sources based on nonlinear mixing of high power, tunable diode lasers and fixed frequency light sources are developed for high resolution, wavelength modulation spectroscopy. These light sources are suited for quantifying key combustion species in flames by high sensitivity $2f$ absorption in the UV and for concurrent $2f$ LIF detection. Results of CH and OH monitoring in laboratory diffusion flames are summarized and approaches to extend these measurements to other combustion species are discussed.

INTRODUCTION

Quantitative mapping of combustion species with good spatial resolution is essential for improved understanding of flame chemical kinetics and fluid dynamics. Laser induced fluorescence (LIF) spectroscopy has been a main staple of combustion diagnostics due to its high detection sensitivity, selectivity, and excellent spatial resolution. However, extracting quantitative species densities from LIF measurements requires careful calibration of geometric factors and laser intensity, as well as modeling of fluorescence quenching by collision partners. This has been challenging even under the most favorable conditions.[1]

Wavelength modulation absorption spectroscopy (WMS) provides quantitative, high sensitivity detection of trace species[2] and is used in a diverse range of applications including combustion studies.[3,4] Minimum detectable absorbances of 10^{-6} have been demonstrated with current-modulated diode lasers.[2-4] This approach provides the integrated absorbance along a line-of-sight optical path, but does not characterize species distributions along the line-of-sight.

LIF detection perpendicular to the laser beam can be used to map relative species densities along the line-of-sight while the integrated absorption available through WMS provides a mathematical constraint on the extraction of quantitative information from the LIF data. Combining line-of-sight absorption with LIF – especially if the measurements are made simultaneously with the same excitation beam – has the potential to yield quantitative, spatially-resolved combustion diagnostics of key intermediates in reactive flow fields.

We have been developing tunable, wavelength modulated UV sources based on nonlinear upconversion of tunable, wavelength modulated high power near-IR diode lasers and high power fixed frequency sources. These sources are appropriate for simultaneous WMS and LIF measurements and have been used to detect CH and OH radicals in laboratory flames. We describe the development of UV sources, the results of CH and OH measurements in flames, and improvements in progress to achieve quantitative, spatially-resolved combustion diagnostics.

CH DETECTION

Figure 1 shows a schematic of the experimental arrangement for CH radical detection. The 852 nm DBR (distributed Bragg reflector) diode laser (120 mW) output was collimated, circularized, and focused into a 7 mm long, A-cut, $KNbO_3$ crystal. The crystal is temperature tuned for non-critical phase matching operation near

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[*] Senior Research Scientist
[†] Senior Research Scientist, Member

The output wavelength of the pump DBR laser diode was tuned by the temperature/diode current settings to the $R_2(8)$ (23,460.81 cm^{-1}) line in the (0,0) band of the CH A $^2\Delta - X ^2\Pi$ transition.[5] In addition to several CH lines in this (0,0) band, the frequency-doubled laser can access some lines in the (1,1) band that would allow optical thermometry in a future study.

Figure 1. Experimental arrangement for a generation of tunable, wavelength modulated 426 nm, where DL (diode laser), FR (Faraday rotator), AP (anamorphic prism), NLC (nonlinear crystal), DP (dispersing prism), x1 (residual 852 nm pump), and x2 (doubled 426 nm) are denoted.

10°C. With 85 mW of near-IR beam coupled into the crystal, 100 μW of 426 nm was produced.

The generated 426 nm beam was propagated along the 4 cm flame front of either an ethylene/air or a methane/air diffusion flame supported on a Wolfhard-Parker slot burner. Figure 2 shows the arrangement for detection of CH radicals by $2f$ WMS absorption and by phase-sensitive LIF.
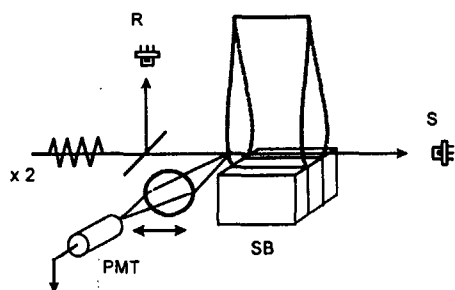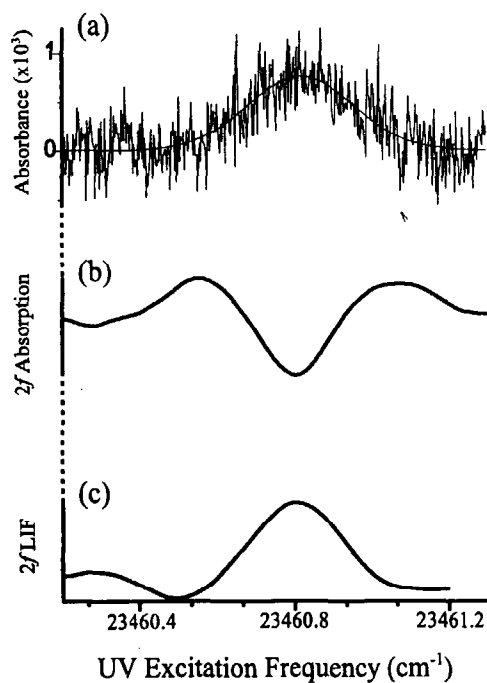

Figure 2. Detection of CH radicals in a laboratory diffusion flame by $2f$ WMS absorption and $2f$ LIF, where R (reference detector), S (signal detector), SB (slot burner), and PMT (photomultiplier tube) on a translation stage are denoted. x2 indicates the frequency doubled diode laser beam.

UV Excitation Frequency (cm^{-1})

Figure 3. Flame spectra of CH acquired from (a) direct absorption, (b) $2f$ WMS absorption, and (c) $2f$ LIF detection.

Figure 3 shows spectra from (a) direct absorption, (b) $2f$ absorption detection, and (c) $2f$ LIF detection at 2 mm above the burner surface in an ethylene/air flame. The peak CH absorbance at this height measured by direct absorption is 7.5×10^{-4} with a peak-to-peak S/N of 2:1. Assuming a uniform 2000 K flame temperature along the 4 cm long flame front, and using reported CH electronic transition moments and rotational transition probabilities,[6] we estimate a peak CH mole fraction of 3 ppm. The $2f$ LIF spectrum shown in Fig. 3(c) is taken with the same detection bandwidth and averaging time as the $2f$ absorption spectrum in Fig. 3(b). The WMS and LIF S/N are excellent and the $2f$ LIF detection scheme is

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effective in removing interference from flame luminescence.

Figure 4 shows CH concentration profiles, constructed from $2f$ absorption spectra for ethylene/air and methane/air diffusion flames with equivalent fuel/air mass ratios. Measurements were made at two different flame heights (2 mm and 4mm) above the burner surface. A zero mm lateral position corresponds to the center of the burner and ± 4 mm are the positions of the fuel/air interfaces at the burner surface. The peak absorbance decreases and shifts further away from the burner center as the height in the flame increases. The peak CH mole fraction in the methane/air flame at 2 mm is 2/3 that in the ethylene/air flame.

These peak CH mole fractions are comparable to the 0.08 ppm to 2 ppm range estimated by Norton and Smyth[7] from LIF measurements on a methane/air diffusion flame using a similar burner and flow rates, but at a 9 mm height. They observe similar trends in CH concentration profiles with change in flame height. The accuracy of these concentration determinations can be improved in the future by incorporating optical thermometry to measure local flame temperatures simultaneously with $2f$ WMS and LIF.
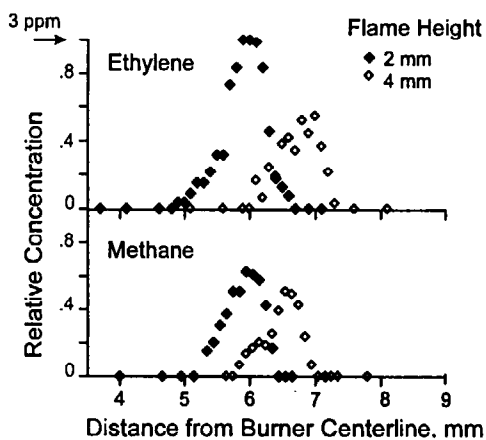

Figure 4. Distribution profile of CH radicals from ethylene/air and methane/air slot burners at two different flame heights.

OH DETECTION

Figure 5 shows the experimental arrangement used to generate a tunable, frequency modulated 308 nm beam by sum frequency mixing. About 1 Watt of 488 nm, single longitudinal mode output from an Ar+ laser was combined
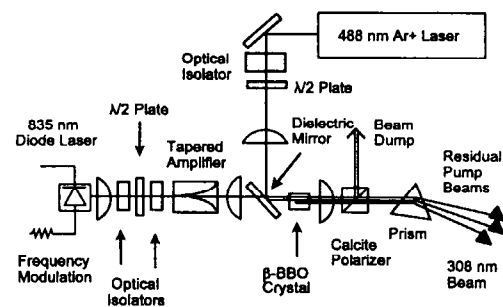

Figure 5. Experimental arrangement for the generation of tunable, wavelength modulated 308 nm for OH detection.

with up to 500 mW of tunable 835 nm output in a β-BBO crystal for sum frequency mixing. The 835 nm source consisted of a single mode, tunable and wavelength modulated diode laser oscillator and a power amplifier to increase the output power to ~500 mW. Up to 4 μW of 308 nm was generated under this direct, single pass mixing arrangement.

The generated 308 nm beam was used to probe OH radicals in an ethylene/air premixed flame (~10 mm dia.) by accessing the $P_2(4)$ line of the (0,0) band of the A $^2\Sigma^+$ – X $^2\Pi$ transition.[8] The detection arrangement is shown in Figure 6. The flame was translated vertically at 10 mm intervals and the corresponding $2f$ absorption spectrum was acquired at each flame height in a line-of-sight
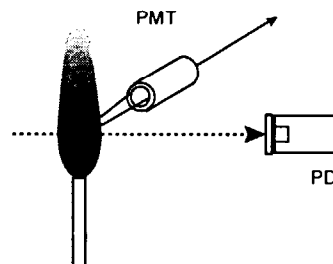

Figure 6. Experimental detection of OH in a laboratory ethylene/air premixed flame, where PMT (photomultiplier tube) and PD (photo diode detector) are indicated.

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configuration using a photdiode. A 2 mm-long column of LIF was collected at 90°with a f/3 UV collection optic. An 1/8-meter monochromator was used as a bandpass filter, and the LIF was detected with a PMT.

Figure 7 shows the resulting $2f$ WMS absorption spectra of the OH $P_2(4)$ line at the (0,0) band of the A $^2\Sigma^+$ - X $^2\Pi$ transition at 5 different flame heights.

Figure 8 shows the corresponding peak absorbances at the 5 flame heights. The absorbance ranges from 0.16 to 0.03. Also shown is the estimated OH mole fraction of 0.5% at a 10 mm flame height when we assume a 2000 K uniform flame temperature across the 10 mm effective path length.
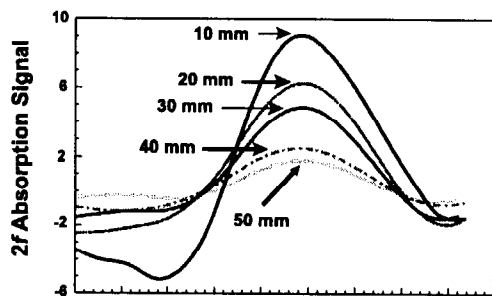

Figure 7. The $2f$ WMS absorption spectra of OH $P_2(4)$ line at (0,0) band of A $^2\Sigma^+$ - X $^2\Pi$ transition acquired at 5 different flame heights.
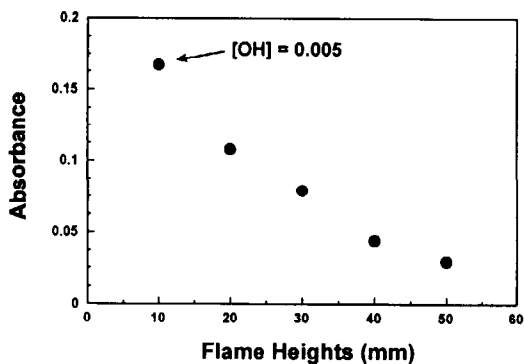

Figure 8. The peak absorbance of $P_2(4)$ OH line at 5 different flame heights, and the extracted OH mole fraction of 0.5% when assumed an uniform 2000 K temperature and 10 mm effective length is for the absorption data at 10 mm flame height.

Additional experiments are underway to quantify the OH density distribution along the beam path by (1) spatially-resolved phase-sensitive OH LIF measurements, and (2) spatially-resolved optical thermometry by accessing two or more OH transitions with different lower quantum states. These measurements will be used to map species distributions and to quantify the line-of-sight $2f$ absorption measurements. After further validations of the technique in laboratory flames, OH measurements in a jet turbine combustor facility will take place.

DETECTION OF OTHER SPECIES

We have also looked into the feasibility of applying these approaches to other species. Table I lists the absorption wavelength ranges needed to access strong transitions for the important combustion radicals C_2, CN and NH in addition to CH and OH. Also listed are the pump wavelengths needed to generate the absorption wavelengths by either second harmonic generation (×2) or sum frequency mixing (SFM). The conditions used for CH and OH in our studies to date are underlined.

Table I Generation of UV wavelengths for species.

Species	Transition	Absorption Wavelength (nm)	Pump Wavelength (nm)
CH	(0,0) A$^2\Delta$ ← X$^2\Pi$	420 - 440	(×2) 840 - 880
C_2	d$^3\Pi_g$ ← a $^3\Pi_u$ (Swan band)	438 - 516	(×2) 876 - 1032
CN	B $^2\Sigma^+$ ← X $2\Sigma^+$	359 - 422	(×2) 718 - 844
NH	A $^3\Pi$ ← X $^3\Sigma^-$	320 - 340	(×2) 640 - 680
OH	(0,0) A $^2\Pi^+$ ← X $^2\Pi$	306 - 330	(×2) 612 - 660 (SFM) 488 + 852
	(1,0) A $^2\Pi^+$ ← X $^2\Pi$	280 - 287	(×3) 840 - 860 (SFM) 488 + 680

The most important factors governing the detection of these radicals are laser wavelength availability, laser power sufficient for non-linear conversion, absorption line strengths, and concentrations. All of these radicals have strong to very strong transition strengths and should be detectable by WMS and LIF if the appropriate wavelengths and powers are available. Because of lower concentrations, the nitrogen containing radicals will be more challenging to detect.

At this time, the requisite wavelengths are available from currently marketed diode lasers. However, in many cases sufficient power is lacking. Given the rapid development of new diode lasers and power amplifiers in the past few years, we believe this approach to flame CH and OH radical detection and mapping will soon be applied to other species.

SUMMARY AND CONCLUSIONS

Tunable, wavelength modulated UV sources based on nonlinear upconversion of high power diode lasers and fixed frequency high power lasers have been developed. Laboratory flame measurements of CH and OH radicals by $2f$ absorption detection show high detection sensitivity. Concentration profiles have been extracted by assuming a uniform, fixed temperature along the absorption path. Further experimental refinements including optical thermometry and determining the effective path lengths and concentration profiles from spatially-resolved LIF measurement are underway. The prospects of detecting other combustion species at shorter UV wavelengths are excellent as developments in higher power diode lasers and fiber amplifiers progress.

ACKNOWLEDGMENTS

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