# Non-Intrusive Combustion Diagnostics

Edited by

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#### Non-Intrusive Combustion Diagnostics

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### **PREFACE**

In the last ten to fifteen years, there have been substantial developments in non-intrusive combustion diagnostic techniques. Advances in diagnostics have been due largely to the rapid development of laser-based optical techniques, computer-coupled data acquisition systems, and digital image capture and analysis. The non-intrusive character of these methods is essential to prevent perturbation of the flow systems under study, and thus provide more realistic and accurate measurements than those provided by intrusive techniques. Advanced diagnostic techniques have been applied to various combustion and propulsion systems for measurements of concentrations of chemical species, temperatures of gaseous mixtures and particles, the velocity field of chemically reacting flows, the regression rate of condensed phase materials, etc. Using advanced non-intrusive diagnostic techniques, engineers and scientists are much better equipped to study combustion, propulsion, and environmentally related problems.

Experimental measurements are needed for a variety of purposes, including achieving a better understanding of flame and flow field structures in order to improve the design of existing combustion and propulsion systems, validating theoretical models and numerical codes, studying the formation mechanism of undesirable species in emission control, and conducting performance evaluations of combustion systems.

Although the design of many modern propulsion systems leans toward calculation and simulation rather than extensive hardware fabrication and testing, diagnostic techniques are still very important. Indeed, the rise in Computational Fluid Dynamics (CFD) is beginning to allow evaluation of complex flow and combustion systems over a wide range of design parameters more quickly, inexpensively, and thoroughly than can be accomplished with actual hardware tests. Reactive flows including multi-step kinetic considerations, such as supersonic combustion in ramjets or solid-propellant combustion in rocket motors, have become computationally tractable. However, these computational models often require adequate input of kinetic information in the form of submodels, which are based upon specific measurements or basic understanding of the chemical and physical mechanisms derived from experimental observations. In particular, experimental validation of computed results will be necessary to prove the predictive capability of theoretical models and computer codes before they are used for guiding actual designs. Advanced non-intrusive combustion diagnostics are an excellent way to provide required input and measured data for comparison with CFD results. Many modern diagnostic techniques allow multiparameter measurements which further improve the validation of CFD results. These multiparameter diagnostic measurements are even more valuable for some systems which, due to complicated geometries or operating conditions, are beyond the ability of current CFD calculations. For such systems, improvement of the design beyond simple trial and error can be enhanced greatly by non-intrusive diagnostic measurements of properties of chemically reacting flow fields.

Based upon strong interest and the need for greater communication on the topic of Non-Intrusive Combustion Diagnostics, the Third International Symposium on Special Topics in Chemical Propulsion was held May 10-14, 1993, in Scheveningen, The Netherlands. The objectives of this symposium were (1) to promote communication between researchers, instrument users, and manufacturers regarding the merits and limitations of advanced non-intrusive diagnostic instruments, (2) to compare different types of combustion diagnostic techniques in terms of their capability for specific property measurements in combustion environments associated with burning of various types of propellants and fuels in either liquid, gas or solid phases, (3) to promote the exchange of information, and (4) to encourage the development of new combustion diagnostic methods for chemical propulsion systems.

One hundred and twenty-five researchers from seventeen countries participated in the symposium. Eighty-one presentations were given, including eight invited talks, forty-nine oral papers, and twenty-four posters. The eight invited speakers were Professor Ronald K. Hanson of Stanford University, Professor Franz Durst of the University of Erlangen-Nürnberg, Dr. John Stufflebeam of the United Technologies Research Center, Dr. Katharina Kohse-Höinghaus of the DLR-Institut für Physikalische Chemie der Verbrennung, Professor Vladmir E. Zarko of the Russian Academy of Sciences, Professor Thomas Brill

of the University of Delaware, Dr. Timothy P. Parr of the U.S. Naval Air Warfare Center, and Professor Robert W. Dibble of the University of California at Berkeley.

This edited book is not simply a record of symposium proceedings with papers reviewed based upon extended abstracts. The full manuscript of all papers has been evaluated through a comprehensive review process. Sixty-three of the eighty-one papers presented at the symposium were accepted for publication. We extend our deep appreciation to many specialists who reviewed and selected the manuscripts published in this volume. The accepted papers are grouped in ten chapters covering many topical areas, including (1) Laser-Induced Fluorescence (LIF and PLIF) Techniques, (2) Coherent and Spontaneous Raman Spectroscopies, (3) Absorption and Emission Spectroscopies, (4) Holographic and Microwave Interferometries, (5) Particle Diagnostics, (6) X-ray Diagnostics and Image Analyses of Combustion of Liquids and Solids, (7) Diagnostics of Gaseous Reaction Systems, (8) Flow Field Measurements and Visualization, (9) Combustion Diagnostics of Solid Propellants, and (10) Diagnostics in Practical Combustion Systems. Invited papers are presented at the beginning of each chapter. Since a large percentage of the papers employ multiple techniques in combustion diagnostics, it is difficult to group some of them into a specific area. It is recommended that readers interested in multiple measurement techniques scan all chapters. Written questions and comments raised by the audience after paper presentations and replies by the authors are given at the end of each paper.

The symposium was sponsored by TNO Prins Maurits Laboratory of The Netherlands, The Pennsylvania State University, the European Space Agency (ESA), the European Research Office of the U.S. Army, SNPE of France, and the EOARD of the U.S. Air Force. The International Science Foundation, a private charitable foundation established by Mr. George Soros, contributed to a portion of the travel costs for participants from the Commonwealth of Independent States. We would like to thank Dr. Ir. Hans J. Pasman, Dr. Tark Wijchers, Dr. Henk J. Reitsma, Ir. Paul A.O.G. Korting of TNO, and Dr. Herman F.R. Schoeyer of ESA for their help in developing the symposium and for local arrangements, and Mr. Slachmuylders of ESTEC of ESA for delivering the welcoming speech.

We thank the members of the steering committee for providing input into the structure and scope of the meeting, participating in the selection of invited speakers, assisting in the review and selection of abstracts for presentations, chairing sessions, and publicizing the meeting throughout the propulsion community. The volunteer assistance of Barbara Pein and Olivia J. Kuo during the symposium registration is appreciated, and we would like to thank Connie Peters and LaRue Jacobs for their help in maintaining excellent records of manuscripts, review comments, communication documents between various parties, developing the symposium program, and assisting in the preparation of this volume. Special thanks are given to Ruth Fergus for her assistance in the English editing of the manuscripts and for establishing a uniform format and style.

We believe that this book provides an excellent introduction to the state-of-the-art technology in non-intrusive combustion diagnostics for propulsion systems to researchers wishing to adopt the methods directly, to those interested in assessing the accuracy, advantages, and limitations of specific techniques, and to those seeking a starting point for new ideas toward advances in combustion diagnostics. This volume should serve as a basis for the future development of advanced instrumentation and diagnostic techniques with higher spatial and temporal resolutions for use in many challenging combustion environments and propulsion systems.

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## **CHAPTER 1:**

LASER-INDUCED FLUORESCENCE (LIF AND PLIF) TECHNIQUES

## QUANTITATIVE ABSORPTION AND FLUORESCENCE DIAGNOSTICS IN COMBUSTION SYSTEMS

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#### **ABSTRACT**

Laser-based diagnostic tecliniques, developed primarily within the combustion community, offer considerable promise for nonintrusive measurements in reactive gaseous flows and propulsion systems. In this paper we overview three diagnostic methods under development at Stanford University: spectrally resolved line-of-sight absorption, conducted with wavelength-modulated semiconductor diode laser and ring dye laser sources; spectrally resolved single-point laser-induced fluorescence, using a rapid-tuning ring dye laser; and planar laser-induced fluorescence, conducted with tunable pulsed dye laser source and intensified CCD array cameras. These methods have unique capabilities for nonintrusive measurements of flowfield properties such as temperature, species concentration, velocity, density and pressure, as well as quantities derived from these parameters such as mass and momentum fluxes. Species monitored in current studies include NO, OH, O<sub>2</sub> and H<sub>2</sub>O.

#### 1. BACKGROUND

Over the past two decades, se cral promising laser-based diagnostics have been proposed for nonintrusive measurements in gaseous flows. Interest in combustion, in particular, has driven the development of these advanced measurement techniques. Linear methods, such as absorption and laser-induced fluorescence, are especially attractive owing to their signal strength, species specificity, and relative simplicity of equipment and data interpretation. Equally important is the fact that variations of absorption and fluorescence have been identified for sensing several flowfield parameters of interest, including species concentrations, temperature, velocity, pressure and density.

Laser-induced fluorescence (LIF), originally developed as a single-point diagnostic but extended in 1982 to simultaneous multipoint measurements in a plane of combustion gases, offers the important attributes of spatial and temporal resolution, assuming use of a pulsed laser source. The planar LIF method (known as PLIF) has been rapidly accepted by the combustion community owing to its value for visualizing complex reacting flowfields. (See Refs. 1 and 2 for an overview.) For example, PLIF images of OH produced in a diffusion flame serve to locate the instantaneous position of flame zones and combustion products; and PLIF images of a tracer compound can be used to quantify the

extent of mixing in turbulent jets. During the past few years, as activity in supersonic flow and scramjet combustors has been rekindled, there has also been considerable interest in developing PLIF as a diagnostic for aerodynamic and propulsion applications. The ability of the PLIF method to capture data for many flowfield points simultaneously is particularly relevant to applications in short-duration flow facilities. It should be noted, however, that LIF and PLIF suffer one particular potential disadvantage, namely that conversion of fluorescence data to absolute species concentrations may be handicapped by various uncertainties, especially in the collisional quenching rate. Much of the current research on PLIF is aimed at developing measurement strategies which avoid these difficulties. Fortunately, good progress has been made for some variables, for example to measure temperature through the ratio of two PLIF images, and to measure velocity through PLIF monitoring of Doppler-shifted absorption. By using signal differences and ratios, many of the problems of unknown quench rates are eliminated. Another limitation of LIF and PLIF, when executed in the usual manner with spectrally broad pulsed lasers, is that an individual measurement typically yields, at best, only one flowfield property. In most cases the fluorescence signal is a combined function of temperature, density and mixture composition, and possibly velocity. Signal interpretation can thus be complex, leading to a requirement for multiple measurements.

Recently, methods based on spectrally resolved absorption lineshapes, using either line-of-sight (LOS) absorption or single-point LIF detection, have emerged as strategies which avoid some of the problems of PLIF and also enable simultaneous determination of multiple flowfield properties.<sup>3-11</sup> Spectrally resolved absorption methods using cw laser sources complement pulsed laser techniques in two important ways: (1) cw absorption can be highly quantitative, easily interpreted, and free of calibration uncertainties; and (2) measurements can be made at high repetition rates or even continuously. Of course absorption is usually a line-of-sight method and thus provides poor spatial resolution, but extension of absorption to multiple paths can relieve this deficiency somewhat. Alternatively, single-point fluorescence detection of spectrally resolved absorption can be used in some cases to provide spatial resolution while retaining most of the advantages of the absorption method.<sup>4,6,11</sup> The key limitation to this strategy, of course, is that the signal levels may be weak. The concept is therefore limited to cases with sufficiently high absorption and fluorescence yields.

Another potential advantage of cw laser absorption worth noting is that the low power levels required are compatible with the use of optical fibers, and hence remote location of laser sources becomes feasible. In the case of semiconductor diode laser sources, <sup>7-10</sup> laser absorption also offers high promise for flight applications as well as ground testing, since these sources are compact, rugged, and economical, and they have low power requirements. We anticipate that packaged, stand-alone instruments based on these lasers and spectroscopic principles are likely to have substantial impact in basic and applied studies of combustion and propulsion flows in the future.

In the following, we provide a brief overview of three current research efforts which represent the ideas introduced above. More detailed descriptions may be found in the publications cited in the Reference section of this paper.

## 2. LINE-OF-SIGHT ABSORPTION MEASUREMENTS USING RAPIDLY TUNABLE CW LASERS

Over the past 15 years, our group at Stanford has been active in developing laser absorption diagnostics based on several rapid-scanning narrow-linewidth laser sources, including infrared-emitting lead-salt diode lasers, UV/visible ring dye lasers, and most recently, near-infrared semiconductor diode lasers. The composition of lead-salt diode

lasers can be tailored to enable emission in the spectral range 3.3–30 microns, which provides overlaps with the IR-active vibrational bands of many combustion species; previous applications include measurements of CO, <sup>12-13</sup> NO<sup>14</sup> and HCN. <sup>15</sup> Ring dye lasers, on the other hand, emit in the range 200-800 nm, which enables use of the stronger absorption bands associated with electronic transitions; species monitored include OH, <sup>16,17</sup> NH, <sup>18</sup> NH<sub>2</sub>, <sup>19</sup> NCO, <sup>20</sup> CH, <sup>21</sup> CN<sup>22</sup> and NO (see below). Near-infrared diode lasers, which are still developing rapidly, presently emit in selected spectral windows from about 650 nm to 1.6 microns. These lasers offer high potential for diagnostic applications owing to their compact size, low cost and compatibility with fiberoptic transmission; we have recently reported results for O<sub>2</sub><sup>7,8</sup> and H<sub>2</sub>O<sup>9,10</sup> using these lasers at 760 nm and 1.385 microns, respectively, and for O-atoms<sup>23</sup> at very high temperatures using a laser at 777 nm.

The elements of high-resolution absorption spectroscopy via wavelength modulation are similar for each of the lasers mentioned above. For brevity, we will focus our discussion here on recent work carried out with a fast-scanning ring dye laser. This laser is a modified version<sup>24</sup> of a commercial ring dye laser, which has been further customized to include an intracavity frequency-doubling crystal (BBO) that extends the operational wavelength range down to 210 nm. The resulting system allows narrow-linewidth scans over a few cm<sup>-1</sup> at repetition rates of 4 kHz; output power in the UV is about 1 milliwatt with 6 W of all-lines UV pump power from an argon ion laser.

Our primary research objective has been to investigate diagnostics strategies, based on spectrally resolved lineshape measurements, for simultaneously monitoring several gasdynamics parameters. The central idea is to scan the laser over a spectral region encompassing two (or more) absorption lines, and then to interrogate the line-of-sight data via a Beer's law analysis to infer: (1) temperature from the ratio of absorption signals for the two lines, assuming a Boltzmann distribution in the absorbing states; (2) pressure from either the collision-broadened linewidth (found with a Voigt fit to the lineshape data) or the absorption level (if the species mol fraction is known); and (3) velocity from the Doppler-shifted position of the absorption lines from that of a static sample. Density can be calculated from the ideal gas law once pressure and temperature are known, and derived quantities such as the mass and momentum fluxes can be calculated from the known density and velocity. Of course, absorption data can also be used for the more limited purpose of determining species concentration, but here our goal was to explore the use of spectrally resolved measurements for simultaneous monitoring of multiple parameters.

We have conducted many of our experiments in a shock tube, since this provides a convenient means of generating a controlled, high-speed flow of gases with precisely known conditions that can be varied over a wide range. Species studied include NO<sup>5</sup> and OH.<sup>3</sup> Nitric oxide is of particular interest because it is both a pollutant in combustion exhausts, and its presence in the flows of many ground-based propulsion facilities provides an opportunity to perform spectroscopic diagnostics on a naturally occurring tracer. The transitions usually studied are in the (0,0) band of the A $\leftarrow$ X system of NO. The experimental arrangement is indicated in Fig. 1. Note that UV laser output is divided into three components. One passes unattenuated to detector A ( $I_0$ ), another passes perpendicularly through the shock tube to detector B ( $I_{90}$ ), and the third beam passes through the shock tube at  $\theta = 60^{\circ}$  with respect to the axis and onto detector C ( $I_{60}$ ). The scanning laser frequency is tracked by detector D ( $I_{\text{étalon}}$ ), which monitors the recurrent transmission of the residual fundamental beam through a fixed-length étalon. Two absorption signals,  $\Delta I_{90} = I_0 - I_{90}$  and  $\Delta I_{60} = I_0 - I_{60}$ , the incident intensity  $I_0$ , and  $I_{\text{étalon}}$  are continuously recorded as the laser sweeps repetitively over a temperature-sensitive rotational line pair. Typically, four scans are completed within the incident-region test time of ~1 ms during which the flow conditions are relatively uniform both spatially and temporally.

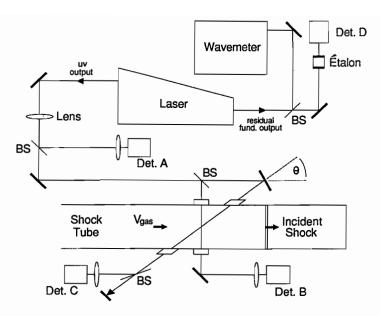


Figure 1. Experimental arrangement for LOS measurements behind incident shocks within a shock tube. The laser output is sent through the shock tube at 90° and  $\theta = 60^{\circ}$  relative to the tube axis. The laser scans rapidly over a spectral feature, and the reference and Doppler-shifted profiles are recorded simultaneously. BS denotes beam splitter.

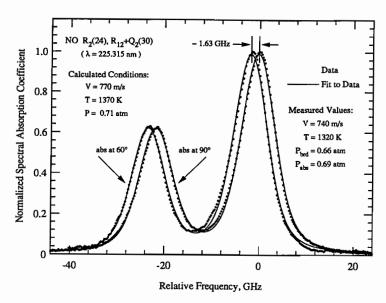


Figure 2. Reduced single-sweep profile of NO LOS data recorded in the incident shock region. As expected, the 60° profile is shifted toward lower frequencies. The measured values are inferred from Voigt fits to the data and compare well with the calculated (theoretical) values. From Ref. 5.

From each scan, measurements of velocity, temperature, and pressure are extracted. Using the Beer-Lambert law and information in the étalon trace, the raw absorption data are converted to traces of spectral absorption coefficient versus frequency. Figure 2 shows a sample reduced trace of NO line pair  $R_2(24)$ ,  $Q_2 + R_{12}(30)$  (centered at 225.315 nm) obtained behind an incident shock of M = 3.50 into a mixture of 0.5% NO in Argon. The agreement between the measured post-shock conditions and the theoretical values of 770 m/s, 1370 K, and 0.71 atm is excellent. Lineshapes recorded for the 90° and 60° directions were fit to Voigt profiles, and the measured velocity of 740 m/s corresponds to the measured Doppler shift of 1.63 GHz between the two profiles. For each profile, the measured rotational temperature was obtained from the ratio of line intensities (the area under each "line" representing its intensity) as this reflects the Boltzmann ratio of the absorbing states. Temperatures of 1320 K and 1310 K were measured for the 90° and 60° The pressure was determined through both the measured beams, respectively. contribution of collision broadening (a parameter extracted from the Voigt fit) and the value of peak absorption using the known NO mole fraction. The values of 0.66 atm and 0.69 atm, returned by the broadening and absorption methods, respectively, are in close agreement with the theoretical value of 0.71 atm. Simultaneous measurements of NO velocity, temperature, and pressure were conducted over a wide range of incident-shock conditions, and these results are collected in Figs. 3a - 3c. Agreement between measured and theoretical values was typically within 7%.

When the flow composition is unknown, measured line intensities of absorption data can yield the absorber partial pressure. In instances of known flow composition, the gas density ( $\rho$ ) can be computed through the measured pressure (P) and temperature, along with the ideal equation of state  $\rho = P/(RT)$ . Thus, as in Fig. 3d, simultaneous measurements of V, T, and P can culminate in the measured mass flux, a quantity of intrinsic importance to propulsion. A similar plot for the measured momentum flux ( $\rho V^2$ ) shows equally good agreement with theoretical calculations.

To summarize our findings with rapid-tuning ring dye laser excitation, spectrally resolved absorption enables accurate, high-repetition-rate measurements of multiple flowfield parameters in pulsed flow facilities. The critical limitation of the method is that it yields a spatially averaged result, and hence the technique is of greatest value in flows which are relatively uniform, such as those found in typical subsonic and supersonic wind tunnels and in many propulsion test facilities. Further details of results obtained with NO are given in Ref. 5; similar work to develop gasdynamic diagnostics using OH is reported in Ref. 3. A technique for continuous velocity measurements, wherein the laser frequency is fixed off line center of the absorption feature, has also been investigated.<sup>3</sup>

In related work to develop multi-parameter diagnostics based on spectrally resolved lineshapes, we have used rapid-tuning semiconductor diode lasers to probe  $O_2$  at 760 nm (see Refs. 7 and 8) and  $H_2O$  at 1.386 microns (see Refs. 9 and 10). These species are of obvious importance in propulsion; for example, a diagnostic capable of sensing  $O_2$  could provide a means of monitoring flows at the inlet of a propulsion system, while a sensor for  $H_2O$  would allow monitoring of exhaust gases. In fact, our work was initially motivated by the objective of developing a nonintrusive optical diagnostic for monitoring inlet mass flux and exit plane thrust of supersonic propulsion systems. Although the absorption lines for the bands indicated are relatively weak, the large levels of these species present in combustion and propulsion systems can lead to acceptable signal levels.

As an example of this work, we display here sample results obtained with the  $H_2O$  diagnostic using a closely spaced pair of lines in the  $v_1 + v_3$  combination band. A typical data trace obtained in a shock-heated flow (with an experimental arrangement very similar to that in Fig. 1 except for the use of a diode laser source: is shown in Fig. 4. Here the two lines probed are the  $6_{42} \leftarrow 6_{43}$  and  $3_{13} \leftarrow 3_{12}$  lines ( $J_{KaKc}$  notation is used; the J = 6 line is the stronger of the two) at 1386.390 and 1386.411 nm, respectively. The data

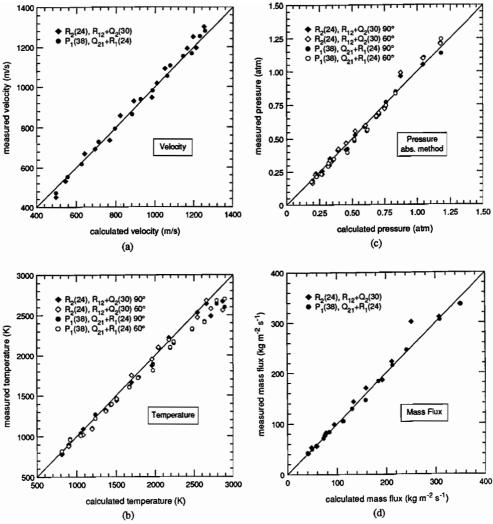


Figure 3. Comparison of NO-based gasdynamic measurements with theoretical values (represented by the solid line): a) axial velocity, b) temperature, c) pressure inferred from the absorption magnitude, d) mass flux. From Ref. 5.

have been plotted as a function of relative frequency (using an etalon record to provide the conversion from time to frequency) and best-fit with a Voigt profile to allow simultaneous determination of temperature (primarily from the ratio of the two peaks) and pressure (primarily from the collision-broadened linewidth). The laser was modulated at a repetition rate of 10 kHz, so the period required for this single-sweep record is 100 microseconds. As indicated on the figure, good agreement is found between the measured and calculated flow conditions for velocity, temperature and pressure. Since the flow is one-dimensional in the shock tube, it is straightforward to convert the measured Doppler shift (between the signals recorded at two angles) to a value for the axial flow velocity. Subsequently, the mass and momentum fluxes can be calculated; for example, the

measured momentum flux  $(\rho V^2)$  is shown in Fig. 5 for the range of experiments conducted, along with a theoretical calculation based on one-dimensional shock wave relations.

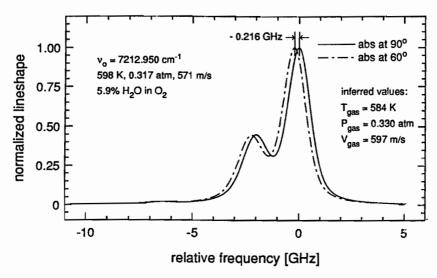


Figure 4. Reduced shock tube absorption data for single scan of the  $6_{42} \leftarrow 6_{43}$  and  $3_{13} \leftarrow 3_{12}$  lines in the  $v_1 + v_3$  band of  $H_2O$ . The gasdynamic parameters inferred from a Voigt profile analysis are indicated along with the values calculated with shock wave theory. From Ref. 10.

Before leaving this topic, we should comment on the utility of UV ring dye laser absorption for sensitive, quantitative species detection using fixed-frequency laser light. In many applications, such as trace gas monitoring, it is only the concentration of a minor species which is of interest. Line-of-sight absorption is an attractive method for such cases owing to both its sensitivity and simplicity. Absorption by strongly allowed electronic transitions, in the visible and UV, is especially attractive, since the absorption coefficients are larger than those of infrared transitions. As an example of achievable detection sensitivity, Fig. 6 presents a graph of the mol fraction of NO which produces 0.1% absorption in a 1 meter path at atmospheric pressure, for two different absorption lines. The detection limit is a function of temperature, but at a typical combustion temperature of 1500 K the limit is well below 1 ppm. The value of 0.1% absorption was selected on the basis of our past experience with laser absorption in laboratory combustion experiments requiring fast temporal response, about 1 microsecond. For static gas measurements, where time-averaging of absorption signals can be used, the detection limit can easily be lowered.

#### 3. SINGLE-POINT LIF USING A RAPID-TUNING RING DYE LASER

In propulsion applications involving nonuniform flow, for example in turbulent combusting flows or in flows with strong three dimensionality, spatially resolved measurements may be required. Furthermore, in most instances, it would be highly desirable to monitor multiple flowfield parameters with one diagnostic, since one variable is insufficient to fully characterize the flow. Unfortunately, previous spatially resolved laser diagnostics typically provide information on only one parameter.

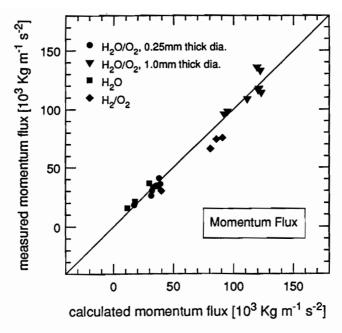


Figure 5. Measured and calculated momentum flux in shock tube using  $\rm H_2O$  absorption diagnostic. From Ref. 10.

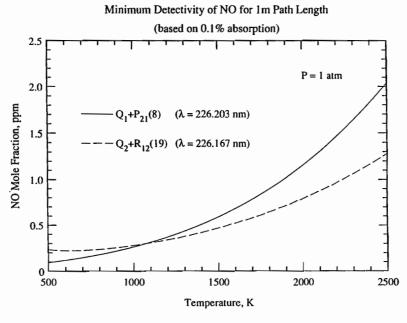


Figure 6. Detection limits of NO using fixed-frequency laser absorption; based on  $L=1\,$  m and 0.1% absorption.

Here we suggest that spectrally resolved absorption lineshapes can be used, in some cases, to provide spatially resolved measurements through the use of fluorescence detection. In single-point LIF strategies, the absorption feature is recorded by collecting and monitoring the broadband fluorescence emanating from a small segment of the illumination beam path. Measurements are thus spatially resolved, since the signal is produced only along that portion of the laser beam which overlaps with the optical train of the collection optics. The basic principles required to infer simultaneous parameters from the spectrally resolved LIF signals are similar to those employed in LOS strategies, except for the fact that the absolute magnitude of the signal, needed to infer the absolute concentration of the species monitored, also depends on the fluorescence yield (the fraction of absorbed photons converted to fluorescence photons). This quantity, the fluorescence yield, may be a complex function of local flowfield properties, and hence may be unknown in some cases. However, a variety of strategies have been developed to deal with this difficulty, including simple calibration under similar but known conditions. A more critical limitation of spectrally resolved LIF is that the signals may be small, since the fluorescence yields are often small. As a result, the development of spectrally resolved single-point LIF using cw laser sources has thus far been limited to electronic transitions (larger Einstein A coefficients, and hence fluorescence yields) accessible with the ring dye laser. Initial work on this diagnostic concept, which was applied to monitor OH in combustion gases, is reported in Ref. 4; more recently, the method has been extended to NO, as discussed in Ref. 11.

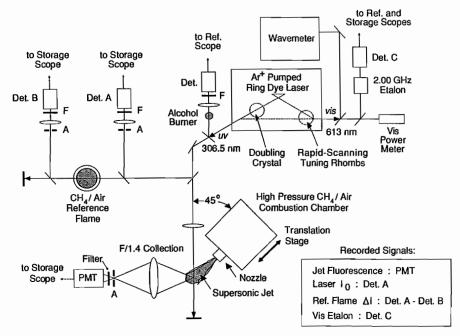


Figure 7. Experimental setup for single-point LIF with a wavelength-modulated laser source. A's, apertures; F's, filters. The solid lines denote the beam paths. Vis, visible.

The experimental arrangement used to monitor OH in a high-speed stream of combustion gases is illustrated in Fig. 7. The laser source is essentially the same as that used in the work described in the previous section on LOS absorption. A pair of fused-silica rhombs

mounted on a single galvanometer in an intracavity-doubled Spectra-Physics 380 ring laser permit the UV output to be swept continuously over a few wave numbers at an effective frequency of 3 kHz. The laser is tuned to scan the OH  $R_1(7)$  and  $R_1(11)$   $A^2\Sigma^+\leftarrow X^2\Pi(0,0)$  line pair at 32,625 cm $^{-1}$ , and the excitation lineshape is recorded in fluorescence. The component of velocity along the beam is obtained from the Doppler shift measured relative to the unshifted lines in a stationary OH source. The temperature is determined from the intensity ratio of the  $R_1(7)$  and  $R_1(11)$  lines, and pressure is inferred from the collisional broadening. The OH concentration can be inferred from the magnitude of the LIF signals, assuming that the fluorescence yield is known.

Measurements were performed in a supersonic (underexpanded), axisymmetric free jet, created by exhausting the products from a high-pressure, stoichiometric CH<sub>4</sub>-air combustion chamber through a 1.6 mm-diameter nozzle into ambient air. Allowing for frictional losses in the nozzle, the jet stagnation pressure was approximately 4 atm. The gas leaves the nozzle at Mach 1 and 2.2 atm and continues to expand isentropically to a pressure less than ambient; an oblique shock then returns the gas to the above ambient pressure. The process is repeated (with decreasing amplitude), resulting in a steady, diamond shock pattern. OH is created as a combustion product and is kinetically frozen during the expansion process at a superequilibrium level of approximately 0.1% (1000 ppm).

The laser provided approximately 15 mW of UV power, of which approximately half reached the jet. The undoubled output of the laser was passed through a fixed, 2.00 GHz visible étalon to provide a frequency marker as the laser was scanned in wavelength. The UV output was focused to less than 0.3 mm in diameter in the jet at an incident angle of 45° relative to the flow. Broadband fluorescence was collected orthogonal to the jet and the beam and was directed through a Schott UG-5 filter onto a photomultipler tube (PMT). A drilled aperture in front of the PMT defined a spatial resolution of 0.33 mm. A portion of the main beam was extracted before it entered the jet and was directed onto a detector (Det. A) to provide a laser-intensity reference signal ( $I_0$ ). A second beam was passed over an atmospheric  $CH_4$ -air burner onto a second detector (Det. B) to provide an unshifted OH absorption signal (I). The beams were sampled by using front surface reflections off fused-silica plates and were balanced optically by varying the beam-splitter angles. The difference signal ( $\Delta I = I_0 - I$ ) was directly recorded. Data were recorded on a four-channel, 400 kHz-bandwidth digital oscilloscope that had the option of waveform averaging so that both single-sweep and sweep-averaged data records could be acquired.

Figure 8 (from Ref. 4) shows normalized line shapes obtained from single-sweep and sweep-averaged traces (40 scans) for a point on the jet centerline at a distance of 2.2 mm (x/D = 1.4) downstream from the nozzle exit. For each data set, the frequency position corresponding to zero shift is determined by a fit to the flame data (these fits indicate a reference flame temperature of 1750 K). The jet fluorescence data are corrected for background scattering and the variation in I<sub>0</sub> with frequency and are fitted to profiles. The profiles are calculated by using shift, temperature, and collisional broadening as fitting parameters, while the integrated area of the profile is fixed to that of the data. The best fit is that which minimizes the integral of the squared difference between the calculated and measured profiles. The Doppler shift is linear with velocity, with a proportionality constant of 306.5 m/sec per gigahertz at the OH wavelength. The shift of 2.9 GHz shown in Fig. 8 corresponds to a velocity of 890 m/sec along the direction of beam propagation (or a centerline velocity of 1260 m/sec). Total uncertainties in the measured velocity component are estimated to be ±90 m/sec for the single-sweep data and ±50 m/sec for the sweep-averaged data. For measurement points off the jet centerline, or generally in flows which are not one-dimensional, it would be necessary to add an additional probe beam at another angle to the flow.

As in previous LOS experiments, the temperature is determined through the Boltzmann intensity ratio of the two lines. The fitting procedure recovers the temperature that best matches the intensity ratio of the recorded data. The single-sweep and sweep-averaged profiles yield temperatures of 1130 and 1080 K, respectively, with fitting uncertainties of  $\pm 150$  and  $\pm 40$  K. The total uncertainties in the measured temperature are estimated to be  $\pm 160$  K for the single-sweep data and  $\pm 60$  K for the sweep-averaged data.

In the jet, Doppler broadening dominates collisional broadening by a factor of 3 to 7. Thus a good signal-to-noise ratio (as well as a good knowledge of temperature) is critical for a pressure measurement. Pressure-broadening coefficients for flame gases at combustion temperatures are reasonably well known, with uncertainties of the order of 15%. We used a correlation of Voigt a (the ratio of collisional to Doppler broadening) =  $0.20P \text{ (atm)} \times 1600/T(\text{K})^{1.3}$  for CH<sub>4</sub>-air combustion products, based on previous research (Ref. 17). From the fits of Fig. 8, pressure values of 0.83 and 0.60 atm are obtained for the single-sweep and sweep-averaged profiles, respectively. Total uncertainties in the measured pressure are estimated to be  $\pm 0.4$  atm for the single-sweep data and  $\pm 0.2$  atm for the sweep-averaged data.

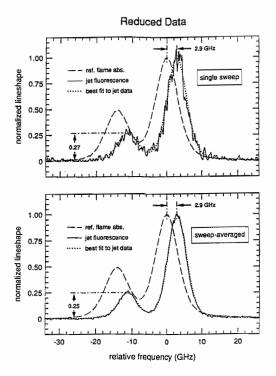


Figure 8. Reduced data for both single-sweep and sweep-averaged scans. The 2.9-GHz line shift corresponds to 890 m/sec. Solid curves, jet fluorescence data; dashed curves, flame absorption data; dotted curves, best fit to jet fluorescence data. From Ref. 4.

To our knowledge, these measurements (reported initially in Ref. 4) represent the first use of fast-scanning LIF to perform Doppler velocimetry in a combustion environment. The ability to measure this critical quantity, as well as to simultaneously determine temperature and pressure, suggests that this laser diagnostic method will find use in a

variety of applications. Furthermore, the capability to obtain data at kHz repetition rates will allow use of the method to study unsteady flows.

Recently we have extended the method of rapid-wavelength-modulation LIF to NO, using the cw ring dye laser system noted in the previous section to access the 225 nm region of the (0,0) band of the A←X system of NO. Details of that work are given in Ref. 11.

#### 4. PLANAR LASER-INDUCED FLUORESCENCE IMAGING

We turn now to our third diagnostics strategy, planar laser-induced fluorescence, known as PLIF, which offers the ability to simultaneously monitor flowfields at a large number of measurement locations. PLIF has developed rapidly from its initial demonstration as a combustion diagnostic in 1982;<sup>25,26</sup> see Refs. 1 and 2 for reviews of PLIF and a detailed treatment of PLIF theory and experimental considerations. Much of the current research on PLIF is concerned with: rendering image data quantitative; extending the concept to new species and flowfield parameters; improving the performance of the equipment, particularly the lasers and detectors; improving analysis of the images through computer-based image processing; and implementation of PLIF systems in propulsion research and testing facilities.

The experimental arrangement employed in one of our current PLIF studies is illustrated in Fig. 9. Sheets of laser light, from two pulsed laser sources, are propagated through the flowfield of interest. If each laser is tuned in wavelength to match an allowed absorption transition of the species to be probed, then a fraction of the light absorbed at each region (point) in the illumination plane is converted to fluorescence which is emitted into all directions. A portion of this light can be collected with lenses (or mirrors) and focussed onto the front face of an intensified solid-state detector array. For most current work, a single-stage microchannel plate (MCP) intensifier is used together with either a photodiode or CCD detector array. The intensifier is time-gated so that light is only collected during a brief period (typically one microsecond or less), to minimize the effects of extraneous emission or scattered light. Current detector arrays have greater than 10<sup>5</sup> pixels, so that each PLIF image is a record of fluorescence emission from a large number of flowfield points in the illumination plane.

In effect, the PLIF signal is a measure of the light absorbed at each flowfield point, modified by the local fluorescence yield, which is the fraction of absorbed light converted to fluorescence emission. Unfortunately, the PLIF signal can be a complex function of the absorbing species concentration, temperature, pressure and velocity (through the Doppler shift of absorption lines for moving gases). Various strategies have been developed to cope with this problem and to relate the PLIF signals to flowfield properties of interest such as species concentration, temperature, 27,28 pressure, density and velocity; see Refs. 2 and 30 for recent overviews. Here we will summarize one of our current projects aimed at developing quantitative imaging of temperature in a flowfield comprised of a jet in supersonic crossflow; this flow is representative of mixing and combustion of a fuel jet in a scramjet combustor. Specifically, the strategy being pursued (known as two-line PLIF) is based on sequential excitation of two absorption transitions of NO (requires two lasers), with the ratio of the two PLIF signals being used to infer temperature. A more complete description of this work may be found in Ref. 28.

The two-line technique used here is based on the ratio of fluorescence signals obtained following sequential laser excitation of two rovibronic transitions, originating from different rotational states in the  $A^2\Sigma^+\leftarrow X^2\Pi(0,0)$  band of NO. This strategy is particularly useful in flows which are compressible or varying in composition, because in taking the ratio of fluorescence signals, the dependence on number density, absorbing species mole fraction, and collisional quenching is removed.

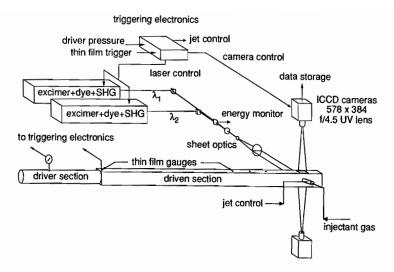


Figure 9. Schematic of the experimental facility for two-line PLIF imaging of temperature in supersonic flows.

NO is an attractive probe species for this application because it is naturally present in the freestream of many high enthalpy combustion and propulsion facilities and can be easily seeded into fuel jets. In reacting flows, the NO may sensitize ignition and may be partially consumed by the combustion process; however, assuming the surviving NO is sufficient for fluorescence detection, accurate two-line temperature measurements are still possible because the NO mole fraction dependence cancels in the signal ratio. Additional attributes of NO as a flow tracer should be noted: (1) it provides a relatively strong absorption cross section and good fluorescence efficiency, which is crucial for imaging applications; (2) it exhibits strong nonresonant fluorescence which, when combined with spectral filtering, can alleviate potential problems with radiative trapping and laser scattering; (3) it does not exhibit J-dependent radiative lifetimes or quenching rates that can lead to systematic measurement errors; and (4) its spectroscopy is well characterized, which facilitates modelling and interpretation of the fluorescence signal.

The two-line method generally requires the use of two lasers and two cameras for instantaneous measurements. In these experiments, the lasers are tuned to different transitions and are fired sequentially, with a delay that is sufficient to temporally separate the respective fluorescence signal decays but less than that which allows blurring of the images due to flow motion. The cameras are gated so that each camera integrates the fluorescence signal induced from only one of the lasers.

For weak laser excitation, the temperature is related to the fluorescence ratio R<sub>12</sub>, by

$$R_{12} \equiv S_{fi}/S_{f2} = C_{12} \exp\{-\Delta \varepsilon_{12}/kT\}$$

where  $S_f$  is the temporally integrated fluorescence signal; the subscripts 1 and 2 refer to the values associated with the respective fluorescence images;  $\Delta \epsilon_{12}$  is the energy difference between the initial absorbing states; k is the Boltzmann constant; T is the gas kinetic/rotational temperature; and  $C_{12}$  is a constant, which is dependent upon spectroscopic and experimental parameters, including the laser pulse energies. The

constant,  $C_{12}$ , can be determined by independent calibration or from the signal ratio directly (in situ) if the temperature is accurately known at some location within the image. Note that in writing this equation, we have assumed that the respective fluorescence yields cancel in the ratio. This is a valid assumption for these measurements because the quenching cross sections, fluorescence lifetimes and fluorescence branching ratios for the  $A^2\Sigma^+$ , v'=0 state of NO are known to be insensitive to rotational quantum number. Differences in the overlap integrals of the absorption and laser lineshape functions for the two lines probed are also neglected.

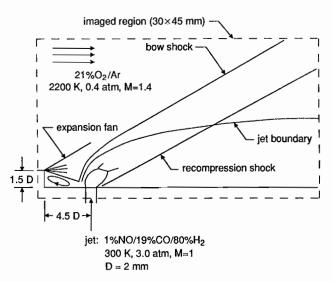


Figure 10. Schematic of the jet-in-crossflow flowfield showing the nominal flow conditions.

The scramjet model flowfield examined here is shown schematically in Fig. 10. The crossflow is supersonic and is turned through a Prandtl-Meyer expansion fan as it passes over the rearward facing step. The flow separates as it passes over the step and forms a recirculation zone just downstream of the step. Fuel is injected sonically through a circular orifice (diameter = D) located 4.5D downstream of a rearward facing step (height The fuel jet is underexpanded and results in the characteristic barrel shock = 1.5D). The barrel shock is swept over due to the momentum of the oncoming crossflow. A bow shock is formed upstream of the fuel jet because the jet acts as an obstruction to the crossflow. Downstream of the fuel jet, the crossflow is turned parallel to the wall by a recompression shock wave. The nominal flowfield conditions for this experiment are listed on the figure and are representative of the inlet conditions for a scramiet combustor. The temperatures and pressures noted in the figure are the static values of the freestream and the stagnation values of the jet gas. Carbon monoxide (CO) was added to the fuel jet to reduce the fluorescence lifetime of NO. This was necessary to ensure adequate temporal resolution of the measurements in this high-speed flowfield. The relatively high freestream temperature (2200 K) was selected to ensure autoignition of the fuel and oxidizer within the short flow time of the observation window.

The lasers used in these experiments were XeC1 excimer-pumped dye lasers (Lambda Physik FL2002 and FL3002). The pump beams at 308 nm had pulse energies of more than 100 mJ with pulse widths of ~20 nsec. Coumarin 450 dye mixtures were used and

the fundamental dye laser beams were frequency-doubled to ~226 nm using BBO crystals. The lasers were tuned to different transitions in the A  $^2\Sigma^+\leftarrow X^2\Pi(0,0)$  band of NO and were fired sequentially with a delay of 250 nsec to temporally separate the fluorescence signals. Including transmission losses through the optics, the pulse energy of each laser sheet (in the test section) was ~0.5 mJ in a spectral bandwidth of ~0.3 cm<sup>-1</sup>.

The broadband fluorescence in the spectral range from 225 to 333 nm was collected at right angles to the plane of illumination with f/4.5 UV Nikkor lenses (105 mm focal length). UG-5 Schott Glass filters (2 mm thick) were used to block elastic laser scattering. The fluorescence was imaged onto two intensified-cooled CCD cameras (Princeton Instruments, with an EEV 578×384 array, 23  $\mu$ m pixels). The imaged region was 30×45 mm. The images were acquired and processed using two IBM-compatible 486 personal computers. Laser pulses were carefully positioned temporally to occur just after the respective intensifier gates were fully on.

Figure 11 shows a typical experimental result for the instantaneous temperature (the precision of the measurement is actually much better than apparent in this gray-scale reproduction). Note that the temperature images are conditioned on the presence of jet fluid (since only the fuel jet contains NO), and hence the images should only be interpreted in regions containing some level of gray. The jet boundary is very easily identified in the temperature field, and is indicative of a very sharp scalar gradient in the shear layer. The plume temperature increases with increasing x/D, due to the warmer freestream mixing with the cool jet. Pockets of relatively cool gas persist within the plume for several jet diameters, but the plume reaches a relatively uniform temperature for x/D≥12. Major pockets of freestream fluid are generally not observed in the lower portion of the plume (adjacent to the wall). The efficient mixing and relatively high temperatures in this boundary layer region make it the most favorable for autoignition and efficient combustion in a reacting flow. This observation is consistent with previous OH imaging measurements in similar combusting flows.<sup>31</sup>

The results obtained for the instantaneous 2-D temperature field via two-line PLIF (see Ref. 28 for details) indicate that shot noise is the dominant source of temperature uncertainty. Single-shot temperature uncertainties varied from a minimum of 5% at low temperatures to as much as 40% at high temperatures for the absorption line pair used. These uncertainties could be reduced, however, by improving the laser pulse energy and efficiency of the collection optics. The good results obtained clearly confirm the potential of PLIF imaging for quantitative thermometry. Other work is in progress in our laboratory to establish quantitative PLIF imaging strategies for velocity<sup>32</sup> and vibrational temperature<sup>33</sup> in high-enthalpy, supersonic flows.

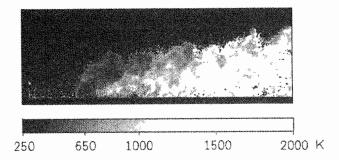


Figure 11. Instantaneous temperature image for a jet in supersonic crossflow.

#### 5. CONCLUSIONS

Each of the three laser techniques described in this paper has advantages and disadvantages. These methods should be viewed as complementary to each other and to other measurement schemes under development in various laboratories. What has become clear in the past few years, however, is that laser-based diagnostics have reached a level of maturity such that they can be employed with confidence to obtain increasingly accurate results in both fundamental and applied research programs on propulsion. Furthermore, these nonintrusive methods can measure flowfield parameters which are, in many cases, inaccessible with the experimental methods utilized in past decades.

#### 6. ACKNOWLEDGEMENTS

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#### COMMENTS

A. Birk, U.S. Army Research Laboratory, USA. Please comment about the applicability of your various diagnostics to high pressure environments (above 100 atm).

Author's Reply. Our experience is with pressures of 1 to 10 atm, but I understand your question. The primary effect of pressure is to broaden the absorption lines, thereby reducing the absorption coefficient at line center (in units of cm<sup>-1</sup>/atm). However, the increased density will still lead to useful signal levels. Another effect is to cause blending

of lines, so that it is more difficult to find isolated lines. My expectation is that both absorption and fluorescence methods will still be useful at pressures of 100 atm and higher, but the strategies will need to be modified. In particular, we will need to tune laser sources over greater ranges in absorption measurements, and in PLIF we will need to account for blended lines in the data analysis. In PLIF, this could actually result in an advantage for measuring some parameters.

M. Lee, DLR-Stuttgart, Germany. The 2-D LIF technique for velocity appears to be useful primarily for high-speed flows. Could you compare the PLIF and absorption techniques for velocimetry, and do you think they are complementary?

You utilized NO seeding fractions (~1%) which may be difficult to handle in larger and more practical systems. In such systems, reduction of this seeding fraction may be necessary. Can you comment on the impact of such reductions on your measurement accuracy, and techniques for improvement of the sensitivity of PLIF measurements?

Author's Reply. Yes, these Doppler-shift methods are primarily useful for high-speed flows. The line-of-sight absorption and PLIF diagnostics are complementary because the former is spatially averaged (and simpler) while the latter gives spatially resolved results (but is much more complex to implement).

The NO seeding level could be reduced somewhat if the laser energy is increased and the f/no of collection optics is reduced. Neither of these parameters was optimized in the current experiments.

M. Sassi, CEDIP/EDF, France. I asked Prof. Hanson to comment on the low velocity limit of Doppler shift measurement methods, especially under high pressure conditions when the line is large and the Doppler shift to linewidth ratio is very small.

Author's Reply. In our earliest work on this subject, Bernhard Hiller was able to demonstrate useful velocity measurements as low as 40 m/sec. These experiments, using iodine as a tracer on an argon laser excitation, were at 0.1-0.2 atm. Your question is a good one, in that you properly recognize the trade off between pressure (affects linewidths) and velocity sensitivity (slope of lineshape function). At high pressures, I expect one should use a broad laser line (broader than linewidth of absorption line), as we do in current supersonic flow studies. There we have been able to resolve about 100 m/sec, so only supersonic flows can be measured with acceptable precision.

J. J. ter Meulen, University of Nÿmegen, the Netherlands. In the two-line strategy examples the excitation takes place toward different rotational or vibrational states. Do you assume that the collisional quenching behaviour of these states is the same?

Author's Reply. Yes. Recent work has shown that there is very little J-dependence to the quench rate for NO at high temperatures. For OH, the error in temperature will be larger, and depends on the rotational states selected. Knowledge of relevant collisional transfer rates and quench rates is now sufficient that reasonable corrections can be made to minimize errors due to J-dependent quench rates.

R. Hönig, Technische Universität München, Germany. Congratulation to your very interesting overview on the different PLIF applications. Especially your turbulent shear flow/mixing studies.

Question 1: Did you ever extend your O<sub>2</sub>-single line thermometry to a 20-line method?

Question 2: Did you try other tracers instead of acetone? What was the influence on the combustion performance?

Author's Reply. With regard to your first question, we have never attempted two-line thermometry with  $O_2$ . The reason is simply that predissociation leads to very low signal levels in  $O_2$ , causing the ratio of two PLIF signals (used to infer temperature) to be very noisy, leading to poor accuracy.

As for your second question, we (and others) have also used biacetyl as a tracer, but acetone is more convenient and suffers less from  $O_2$  quenching. There is some effect on combustion, but we have not analyzed this yet.