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on
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For the Period
October 1, 1990 to October 31, 1991

Submitted by
R. K. Hanson, Principal Investigator

HIGH TEMPERATURE GASDYNAMICS LABORATORY
Mechanical Engineering Department
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Progress is reported for the past year of an interdisciplinary program aimed at establishing advanced optical diagnostic techniques applicable to combustion gases and plasmas, with some emphasis on high speed flows. The primary flowfield parameters of interest are species concentrations (including electrons), temperature, mass density, pressure, and velocity, and quantities derivable from these parameters such as mass flow rate (from the product of density and velocity). The techniques under study are based on laser spectroscopy, particularly laser absorption and laser-induced fluorescence, with the latter capable of providing both single-point and multi-point (2-d and 3-d) measurements. Laser sources include tunable cw lasers (ring dye and semiconductor diode lasers) and tunable pulsed lasers (excimer-pumped dye and narrow-linewidth excimer). The cw lasers are spectrally narrow, allowing study of a new class of techniques based on spectral lineshapes and shifts, while the pulsed lasers provide intense bursts of photons needed for techniques based on light-scattering phenomena. Accomplishments of note include: the first optical measurement of mass flux in high speed air flows; the first applications of tunable semiconductor diode lasers to absorption and fluorescence measurements in high temperature plasmas and supersonic flows, and to the measurement of water vapor in high temperature combustion gases; the first application of the planar laser-induced fluorescence (PLIF) technique to nonequilibrium shock tunnel flows; and further advances in the development of shock tube diagnostics for rate constant measurements of elementary combustion reactions.
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# TABLE OF CONTENTS

<table>
<thead>
<tr>
<th>Section</th>
<th>Title</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.0</td>
<td>INTRODUCTION.</td>
<td>1</td>
</tr>
<tr>
<td>2.0</td>
<td>PROJECT SUMMARIES.</td>
<td>2</td>
</tr>
<tr>
<td>2.1</td>
<td>Plasma Diagnostics.</td>
<td>2</td>
</tr>
<tr>
<td>2.2</td>
<td>Semiconductor Diode Laser Diagnostics for Gasdynamics Measurements</td>
<td>4</td>
</tr>
<tr>
<td>2.3</td>
<td>CW Ring Dye Laser Diagnostics.</td>
<td>11</td>
</tr>
<tr>
<td>2.4</td>
<td>PLIF Imaging in Nonequilibrium Shock Tube Flows</td>
<td>17</td>
</tr>
<tr>
<td>2.5</td>
<td>Shock Tube Diagnostics for Combustion Kinetics.</td>
<td>22</td>
</tr>
<tr>
<td>2.6</td>
<td>Digital Camera for High-Speed Imaging</td>
<td>23</td>
</tr>
<tr>
<td>2.7</td>
<td>High-Resolution PLIF Imaging in Turbulent Jets</td>
<td>26</td>
</tr>
<tr>
<td>2.8</td>
<td>Multiplex Degenerate Four-Wave Mixing</td>
<td>29</td>
</tr>
<tr>
<td>3.0</td>
<td>PRESENTATIONS AND PUBLICATIONS.</td>
<td>32</td>
</tr>
<tr>
<td>3.1</td>
<td>Presentations (10/90 - 10/91)</td>
<td>32</td>
</tr>
<tr>
<td>3.2</td>
<td>Refereed Publications (10/90 - 10/91)</td>
<td>33</td>
</tr>
<tr>
<td>3.3</td>
<td>Technical Reports (10/90 - 10/91)</td>
<td>35</td>
</tr>
<tr>
<td>4.0</td>
<td>PERSONNEL.</td>
<td>37</td>
</tr>
<tr>
<td>4.1</td>
<td>Postdoctoral Research Associates.</td>
<td>37</td>
</tr>
<tr>
<td>4.2</td>
<td>Graduate Research Assistants.</td>
<td>37</td>
</tr>
<tr>
<td>4.3</td>
<td>Ph.D. Degrees Awarded</td>
<td>37</td>
</tr>
<tr>
<td>5.0</td>
<td>SIGNIFICANT INTERACTIONS.</td>
<td>38</td>
</tr>
</tbody>
</table>
1.0 INTRODUCTION

Progress is reported for the past year of an interdisciplinary program aimed at establishing advanced optical diagnostic techniques applicable to combustion gases and plasmas, with some emphasis on high speed flows. The primary flowfield parameters of interest are species concentrations (including electrons), temperature, mass density, pressure, and velocity, and quantities derivable from these parameters such as mass flow rate (from the product of density and velocity). The techniques under study are based on laser spectroscopy, particularly laser absorption and laser-induced fluorescence, with the latter capable of providing both single-point and multi-point (2-d and 3-d) measurements. Laser sources include tunable cw lasers (ring dye and semiconductor diode lasers) and tunable pulsed lasers (excimer-pumped dye and narrow-linewidth excimer). The cw lasers are spectrally narrow, allowing study of a new class of techniques based on spectral lineshapes and shifts, while the pulsed lasers provide intense bursts of photons needed for techniques based on light-scattering phenomena. Accomplishments of note include: the first optical measurement of mass flux in high speed air flows; the first applications of tunable semiconductor diode lasers to absorption and fluorescence measurements in high temperature plasmas and supersonic flows, and to the measurement of water vapor in high temperature combustion gases; the first application of the planar laser-induced fluorescence (PLIF) technique to nonequilibrium shock tunnel flows; and further advances in the development of shock tube diagnostics for rate constant measurements of elementary combustion reactions.
2.0 PROJECT SUMMARIES

Included in this section are summaries of progress in each of eight project areas. Additional descriptions of this work may be found in the publications listed in Sections 3.2 and 3.3. Reprints of these papers are available on request. Personnel involved in these projects are listed in Section 4.0.

2.1 Plasma Diagnostics

During the past year we have continued to explore the use of cw semiconductor diode lasers (GaAlAs) as light sources for absorption and fluorescence diagnostics of temperature and electron number density in laboratory plasmas. These low-cost lasers are evolving rapidly owing to their numerous practical applications (compact disc players, supermarket scanners, etc.), but they also offer exciting prospects for use in scientific applications. With regard to spectroscopic diagnostics, these lasers can be viewed as economical, rugged and compact sources of low power, cw, tunable wavelength light with relatively narrow spectral linewidths. Thus they represent possible replacements for currently employed cw dye lasers. Unfortunately, diode lasers have primarily been developed for use at near-infrared wavelengths (especially 1.3 and 1.5 microns), and only recently have lasers become available at wavelengths below 1 micron where most electronic transitions of interest in atoms and molecules are located. At the present time, diode lasers are available for use in selected wavelength bands down to about 650 nm. In our recent work we have utilized diode lasers which operate in the 800-850 nm range, which conveniently overlaps the strong $4s^3P \rightarrow 4p^3D$ transitions of argon linking the first excited state of argon ($4s$) with the second excited state ($4p$).

The experimental arrangement utilized to study both absorption and fluorescence diagnostic strategies is shown in Fig. 1, together with a sketch of representative data which illustrates the features of the data used to infer plasma properties. The measurements are made in an atmospheric pressure, inductively coupled RF plasma torch assembled in the previous year of this program to provide a controlled, high temperature plasma with convenient optical access. In the experiment, the diode laser is rapidly swept in wavelength across an individual absorption transition of interest, providing a fully resolved record of the absorption lineshape; either line-of-sight absorption or single-point fluorescence recording may be used. The absorption data can be used to infer gas temperature (through Doppler broadening of the line) and electron density (through the Stark-induced shift in the line position). Since the lineshape also is influenced by Stark broadening, the linewidth data can be interpreted to yield a second, independent determination of the electron density. In general, excellent agreement is found between the two measurements of electron density. Inferred temperatures are typically in the range 6000-8000 K, for both absorption and
Spectrally-Resolved Plasma Spectroscopy using Tunable Diode Lasers

- First measurements of spectrally-resolved Stark-affected absorption spectra in plasmas.
- Tunable diode lasers allow first simultaneous measurements of electron number density, temperature, and species concentration in atmospheric pressure plasmas.
- Rapid scanning diode laser allows first high repetition rate measurements in plasmas (>1 kHz).
- Fluorescence detection provides high spatial resolution.

Figure 1. Spectrally-resolved plasma spectroscopy using tunable diode lasers.
fluorescence measurements, thereby illustrating the power of this diagnostic for probing very high temperature gases. Finally, it can be noted that the fractional absorption measured across the width of the plasma can be used to infer the population density in the absorbing excited state and hence a population temperature for this level.

These initial experiments apparently represent the first use of diode lasers in plasma diagnostic schemes. The success achieved in simultaneously determining multiple plasma properties using a single, economical laser source suggests that this is a promising line of research. We therefore plan to continue and to expand our research on this topic in the coming year. Further details of this work may be found in the publications of Baer and Hanson cited in Sections 3.2 and 3.3.

2.2 Semiconductor Diode Laser Diagnostics for Gasdynamics Measurements

In addition to the work described in Sec. 2.1 on plasma diagnostics, we have made good progress in establishing a new class of diagnostic techniques, based on diode laser absorption, which will enable accurate and fast measurements of gasdynamic properties such as velocity, temperature, pressure and density in high speed gas flows. The fundamental concept is the same as developed previously in this laboratory with tunable cw ring dye lasers (see also the following section 2.3), namely to utilize the dependence of spectral lineshapes on the thermodynamic and gasdynamic state parameters of interest. Such a strategy permits simultaneous determination of multiple flowfield variables, and with a measurement repetition rate far in excess of current alternative diagnostic schemes based on pulsed lasers. Tunable diode lasers are attractive and highly promising sources of the narrow-linewidth radiation needed because they are (or will be) economical, rugged and compact, and in addition they can be modulated in wavelength at extremely high rates. At present, diode lasers are available only in specific wavelength windows at wavelengths generally in excess of 750 nm, but the trend toward operation at shorter wavelengths and toward increasing the number of "laser wavelength windows" is clear.

In the past year, our research has been focussed on two related efforts: one involving detection of O₂ with 760 nm radiation, and the other involving H₂O detection with lasers operating near 1.4 microns. These projects are discussed separately below.

Our work with O₂ is now in its second year. In the first year we identified the fortuitous spectral match between recently developed GaAlAs diode lasers which emit near 760 nm and the weakly absorbing A-band (known as the "atmospheric band") of molecular oxygen, and we assembled a bench-top diode laser absorption system to establish optimum modulation and detection strategies. The specific transitions of O₂ utilized are shown in Fig. 2, which is a plot of the fractional absorption which will occur for the various transitions
in the R-branch of the A-band for a 5 meter path of air at room temperature. Note that the region studied in this first effort (see the box in Fig. 2) encompasses two lines, which enables inference of temperature from the ratio of absorption in the two transitions, and that these lines correspond to relatively high rotational quantum number so that they will be useful over a broad range of temperature including combustion conditions. The peak absorption in room air is about 1% per meter. This weak absorption, especially for the short paths found in laboratory test facilities, leads to the requirement for highly sensitive detection schemes. The approach we've chosen involves scanning the laser wavelength (by varying the current through the diode) with a relatively fast ramp waveform superposed with a smaller amplitude, high frequency sinewave at frequency f. The absorption signal is detected at a harmonic of f, usually 2f, to allow discrimination against various forms of noise. The result is a fast, repetitive recording of the second derivative of the absorption spectrum. This 2f spectrum is compared with theoretical results for the spectrum which depend on the gas velocity (produces a Doppler shift in line positions), the gas temperature (depends on the ratio of peaks in the two lines), the gas pressure (influences the linewidths) and the O₂ partial pressure or concentration (influences the peak signal levels).

![Figure 2. Calculated transmission for 5 meters of room air; R-branch of A-band.](image-url)
The experimental arrangement as employed in recent measurements is shown schematically in Fig. 3. In this case the modulation frequency is 10 MHz (detection is at 20 MHz) and the ramp frequency is 10 kHz; thus a complete measurement is made each 100 microseconds. A detailed sketch of the actual optical arrangement employed for shock tube experiments is shown in Fig. 4. Here two beams are passed through the shock tube to enable simultaneous recording of a Doppler-shifted signals and a non-shifted signal. This leads to cancellation of pressure-shift effects and more accurate detection of velocity. A sample data trace is shown in Fig. 5; note that this data sequence allows multiple determinations of the properties for three consecutive stages of shock tube conditions. Example results for velocity are shown in Fig. 6. These results confirm the utility of this new diagnostic to measure air velocity over a wide range of conditions and with good accuracy. To our knowledge, this is the first non-intrusive spectroscopy-based velocity diagnostic demonstrated in high temperature, high-speed air flows. Perhaps more importantly, the ability to infer multiple flow properties leads to a new capability for determining derived flow quantities such as mass flux. An overview of these ideas and their attributes is given in Fig. 7. Details of the strategy to develop a diode laser sensor for monitoring mass flux in supersonic air flows are described in a recent paper (see the papers by L. Philippe and R. Hanson listed in Secs. 3.2 and 3.3).

Our work with tunable diode laser absorption of H$_2$O is more recent. Initially we conducted an analytical study of water vapor absorption bands to identify possible coincidences of H$_2$O bands and diode laser wavelengths. We found that the strongest bands of water, especially for high temperature measurements, were near 1.4 microns, and that these bands are "combination bands", i.e. they involve simultaneous changes in two (or more) vibrational quantum numbers. Although less strong than the "fundamental" vibrational bands at longer wavelengths, the combination bands near 1.4 microns should still lead to absorption signals at least as large as those obtained in monitoring O$_2$. Thus, in principle, the methods being developed for oxygen should carry over to water vapor. This is important, since it would be desirable to identify a single strategy in which O$_2$ could be detected in air inlets of supersonic propulsion systems (ground-based test facilities or flight systems) and H$_2$O measurements could be made in the engine exhausts. Further, in analogy with the use of O$_2$ detection to give air mass flux, one could imagine applying the same approach to infer thrust at a combustor exit plane. The fact that these diode laser strategies are compatible with optical fiber technology holds promise of applying these measurements remotely and with multiple measurement paths to allow more complete mapping of the flow in question.

After identifying the optimum wavelength regions for absorption measurements, and developing the code to calculate the absorption spectra of water at any prescribed conditions,
modulation frequency: 10 MHz - detection at 20 MHz fast scanning rate (up to 30 kHz)

Figure 3. Experimental arrangement for tunable diode laser absorption of O₂.

Figure 4. Details of optical arrangement for shock tube O₂ experiments.
experimental 2f signal (RQ(13,14) line at 760.258 nm; Δν = 2.2 GHz)

- three consecutive stages at the measurement location

\[ P_1, T_1 \rightarrow P_2, T_2 \quad P_2, T_2 \rightarrow P_3, T_3 \]

- \( P_1 = 0.13 \text{ atm}, T_1 = 300 \text{ K} \)
- \( P_2 = 0.92 \text{ atm}, T_2 = 600 \text{ K} \)
- \( P_3 = 3.90 \text{ atm}, T_3 = 930 \text{ K} \)

calculated absorptions: 0.76%, 1.04%, 0.87%

Figure 5. Raw data for shock tube experiment.
Figure 6. Time-resolved velocity results for shock tube experiment.
Semiconductor Diode Laser Detection of O$_2$
Using Wavelength Modulation Spectroscopy

- Simultaneous density, temperature, velocity and mass flux measurements in air flows

![Diagram showing diode laser detection process]

- Temperature: Ratio of Peaks 1 and 2
- Velocity: Doppler Shift of lines
- Pressure: Magnitude of signals 1 and 2
- Density: $\rho = P / r T$
- Mass Flux: $m = \rho \cdot V \cdot Area$

- Sensitive method based on non-intrusive laser absorption at 760nm (A-band of O$_2$)
- Harmonic detection of wavelength modulated laser extends detection limit
- Lasers are economical, rugged, compact and compatible with optical fibers
- Potential for flight instruments (air inlet, engine exhaust)

Figure 7. Overview of diode laser diagnostic for O$_2$ measurements.
we turned our attention to acquiring a diode laser. After an extensive search, we were able to obtain one sample of a laser being developed by a Japanese company which gives tunable single-mode output near 1.5 microns, and we were able to purchase two multimode lasers which operate near 1.35 microns. Although not optimum for high temperature work, these lasers are suitable for testing various wavelength tuning and modulation strategies. In particular, we are exploring a novel external-cavity approach to convert multimode lasers to single mode operation. In brief, this involves removing the front window on the laser diode and installing a miniature partial reflector whose position can be finely adjusted to enhance gain on a single laser mode. If this is successful, then it will be possible to access single-mode laser radiation outside the wavelength range for which commercial single-mode lasers are available and at the wavelengths ultimately needed for optimum sensing of H$_2$O at high temperatures. Successful experiments have now been conducted both to probe H$_2$O vapor in room temperature air and in the postflame region of a flat flame burner. We have built the codes to fit the measured spectra and infer temperature and water vapor concentration. These results provide strong encouragement for the concept we are pursuing. During the next year we hope to extend these benchtop experiments to include detection of velocity, and also to assemble laser systems which function at preferred wavelengths.

### 2.3 CW Ring Dye Laser Diagnostics

Continuous-wave (cw) laser sources offer important advantages for laser diagnostics which have not yet been exploited. In our laboratory, for example, we’ve developed several new diagnostic strategies over the past 9 years based on cw single-mode ring dye lasers. Such lasers provide a continuous, low power source of nearly monochromatic light well suited for probing the details of spectral lineshapes using either absorption (line-of-sight) or single-point fluorescence (spatially resolved) measurements. The ability to fully resolve absorption lineshapes allows determination of several parameters: (1) highly sensitive and quantitative species concentration measurements from the magnitude of the absorption (or fluorescence) signal; (2) temperature from the ratio of signals from different absorption lines; (3) pressure (or density) from the shape (width) of lines; (4) velocity from the Doppler shift of lines relative to the spectral position of lines in a static sample; and (5) various derived quantities, such as mass flux which is given by the product of mass density and velocity. A schematic of this general measurement strategy is given in Fig. 8. It is important to note here that these various quantities can usually be obtained simultaneously, and, since these measurements are made by rapidly scanning in wavelength, the measurement repetition rate can be very high. In fact, some measurement strategies involve fixing the laser wavelength and probing the flow simultaneously from multiple directions, and in this case the measurements are continuous. It should be clear from the above discussion that measurement techniques based on cw lasers offer important capabilities not possible with
Simultaneous Multi-Parameter Measurements of Supersonic Flows

- Non-intrusive method is founded on novel rapid-scanning cw dye laser
- Measurement repetition rate is \(~4\) kHz
- Demonstrated in 1990 as the first high-speed, multi-parameter measurement technique

- Two Configurations:  
  \[\{ \text{Absorption} \rightarrow \text{Integrated, line-of-sight measurements} \]
  \[\text{LIF} \rightarrow \text{Spatially-resolved measurements} \]

- Mass flux computed from \(V\), \(T\), and \(P\) plus equation of state
- Accessible species include \(\text{OH}, \text{NO},\) and \(\text{O}_2\)

Figure 8. Laser wavelength modulation strategy for simultaneous multi-parameter measurements of supersonic flows.
pulsed laser techniques. The price paid, of course, is that cw lasers are low in power and hence are not generally suitable for imaging measurements which yield data for a large number of measurement points simultaneously.

In last year’s annual report, we described three significant accomplishments: (1) the first application of cw dye lasers to perform single-point LIF of OH; this new diagnostic yielded simultaneous measurements of temperature, velocity and pressure in a supersonic combustion flow, all at a repetition rate of 4 kHz; (2) the first application of wavelength modulation spectroscopy in shock tube flows; these absorption measurements, on OH, NO and O₂, yielded accurate simultaneous data for velocity, temperature, pressure, density and species concentration, all at a repetition rate of 4 kHz; and (3) a new fixed-frequency absorption scheme for real-time measurements of velocity in 1-d flows; the measurements were made with OH, NO and O₂, but the concept is general. With these accomplishments behind us, the goal for this year’s research was to extend the single-point LIF concept to NO, since NO is a more broadly useful flow tracer than OH. In particular, NO can be utilized in flows at both low and high temperatures, while OH is only found in combustion flows at high temperatures.

The extension of cw dye laser wavelength modulation spectroscopy to LIF of NO was motivated by the desire to obtain both spatially resolved and high repetition rate data in flows with wide temperature variations. We thus chose a supersonic underexpanded jet as the flow in which to test and validate our approach. A schematic for the experimental arrangement is shown in Fig. 9. A 0.5% NO in N₂ mixture, initially at room temperature and pressure, is expanded through a 3 mm dia nozzle into a vacuum tank with a background pressure of 0.046 atm. The overall pressure ratio of 22 produces centerline velocities up to 700 m/s and temperatures below 100 K prior to the Mach disc. Example data acquired on the jet centerline at an axial location x/D = 0.75, for a pair of NO lines near 226.7 nm, are shown in Fig. 10. The LIF data were taken with the laser illumination at a 60 degree angle to the flow; a reference absorption record for a static sample is also shown, since this is the basis for inferring the Doppler shift of the absorption line at the measurement point. A comparison of measured and predicted values for temperature, pressure, velocity and Mach number is shown in the inset; the agreement is excellent. Comparisons of measured and predicted centerline temperatures and pressures as a function of axial distance are shown in Figs. 11 and 12; again, agreement is excellent.

In summary, we feel that cw dye laser diagnostics based on fast wavelength modulation spectroscopy provide important and unique capability for combustion diagnostics. Although the methods are limited to single-point and line-of-sight approaches at present, the accuracy of the data, the high measurement repetition rates which are possible, and the ability to simultaneously monitor multiple flowfield variables, are all significant advantages over pulsed laser diagnostic strategies.
Figure 9. Schematic for cw dye laser single-point LIF measurements of NO.
Reduced Fluorescence and Reference Absorption Signals of 
NO $Q_2 + R_{12}\,(8)$, $R_2\,(4) \,(226.7 \text{ nm})$ for Measurements at $x / D = 0.75$

**Table**

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<th>Inferred Values</th>
<th>Predicted Values</th>
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</thead>
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<td>156</td>
<td>159</td>
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<tr>
<td>$P, \text{ atm}$</td>
<td>0.117</td>
<td>0.123</td>
</tr>
<tr>
<td>$V, \text{ m/s}$</td>
<td>549</td>
<td>521</td>
</tr>
<tr>
<td>$M$</td>
<td>2.16</td>
<td>2.03</td>
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</tbody>
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Fluorescence ($a = 1.275$)

Ref. Absorption ($a = 0.050$)

$T_{\text{cell}} = 298 \text{ K}$

$P_{\text{cell}} = 0.004 \text{ atm}$

**Figure 10.** Single-sweep single-point LIF data of NO in a free jet.
Figure 11. Comparison of predicted and measured temperatures in a free jet using single-point LIF of NO.

Figure 12. Comparison of predicted and measured pressures in a free jet using single-point LIF of NO.
In addition to the projects described above, we have maintained contact during the past year with the shock tunnel group at NASA Ames which has been implementing a wavelength modulation system to monitor OH concentration and temperature in its 16-inch tunnel. This is a collaborative effort which has led to the transfer of our previously developed diagnostics technology into a hypersonic flow facility of importance in current research on the National Aerospace Plane.

2.4 PLIF Imaging in Nonequilibrium Shock Tube Flows

Nonequilibrium supersonic/hypersonic flows, relevant to current research on scramjets, pose new measurement problems for experimentalists. For example, experiments are often conducted in pulsed flow facilities in which the available measurement time is quite limited, thereby putting a premium on the ability to acquire complete data sets in very short times. In addition, many flows of interest exhibit a high degree of nonequilibrium, requiring experimental methods sensitive to such nonequilibrium effects. PLIF has high potential for dealing with both of these critical problems, in that the data provide population densities in specific quantum states of the species probed at a very large number of flowfield locations. During the past four years we have initiated research which addresses the primary problems inherent in extending PLIF to transient supersonic flows, and these experiments are now yielding important results. Specific accomplishments of the past year, discussed below, include the first applications of PLIF to: (1) shock tunnel flows; and (2) transverse jet injection into a supersonic cross-flow, with and without reaction.

Our experiments have been carried out in a standard pressure-driven shock tube, built three years ago specifically for research on PLIF imaging diagnostics for supersonic flows. During the past year we have added a shock tunnel to the facility to allow study of PLIF in rapidly expanding, high enthalpy flows. Shock tubes and tunnels provide an economical and convenient means of studying nonequilibrium gasdynamic phenomena and of simulating the extreme conditions relevant to advanced air-breathing propulsion systems. The chief limitation of such facilities, of course, is the short run times which can be produced.

The objective in the shock tunnel work is to identify and study limiting aspects of PLIF imaging for nonequilibrium hypersonic flows. In particular, we wish to establish optimized variations of PLIF which can be used to image velocity, temperature and species concentrations in flows with either vibrational or chemical nonequilibrium. We've focussed work thus far on vibrational nonequilibrium, since the theory needed to compute such flows is more manageable, and we've employed a simple free jet geometry to provide easy optical access to a flow with a wide range of gasdynamic parameters. A schematic of our flow and optical arrangement, and an initial single-shot PLIF image of radial velocity, appears in Fig. 13. The nozzle has a diameter of 2 mm, and for the experiment shown the stagnation
Single-Shot Velocity Imaging in a Hypersonic Shock Tunnel

- Molecular velocity measurement based on Doppler-shifted absorption of NO

![Shock Tunnel Facility](image)

- First application of PLIF to hypersonic and shock tunnel flows
- No particle seeding required
- Can be extended to provide measurement of two velocity components
- Potential for simultaneous temperature and density measurements

Figure 13. Optical arrangement and initial PLIF data for radial velocity in a shock tunnel free jet.
conditions are 2270 K and 2.84 atm, and the gas is an NO/N₂ mixture (proportions are 5:95). The overall pressure ratio (stagnation to ambient) is 108, which produces a jet with an extremely wide range of flow conditions; e.g., the centerline properties just upstream of the Mach disc (located at x/D = 7.0) are: T = 190 K, P = 0.3 torr, and M = 7.5. Thus the temperature varies by a factor of about 12 and the pressure by more than 7000 from the stagnation region to the region just upstream of the Mach disc! Such broad variations provide a stringent test of any diagnostic method’s dynamic range, and also provide a means of generating various types of nonequilibrium. For example, at some point in the flow vibrational freezing will occur, meaning that the vibrational temperature will separate from the rotational/translational temperature; and ultimately, when the pressure drops to a torr or so, rotational nonequilibrium will occur. At present we are focusing on the issue of vibrational nonequilibrium, and we have built (see past progress reports and the publications of J. Palmer cited in Secs. 3.2 and 3.3) a method of characteristics code to provide predictive capability for the flowfield with finite vibrational relaxation rates. Through comparisons with such calculations we hope to optimize and verify our diagnostics strategies for extracting (from PLIF images) the multiple temperatures, velocity (radial and axial) and other flowfield variables which are regarded as important. Although the comparisons between measured and computed velocity are still in the early stages, agreement appears to be good. Perhaps most importantly, we’ve learned in this work that the spectral shape of the laser pulse, on a pulse-to-pulse basis, is critical to accurate velocity measurements, and accordingly we’ve begun to assemble a new diagnostic for monitoring the critical laser properties on a shot-to-shot basis. We believe that this issue of laser lineshape was overlooked in previous work (in our laboratory and elsewhere), and that our study of the problem will be essential to the eventual development of quantitative PLIF imaging techniques.

At the present time, we are making our first attempt to image rotational temperature in this same flow. The approach is based on exciting, sequentially, two separate rotational states, and then forming the ratio of PLIF signal levels at each flowfield point. This ratio is proportional to the relative population density in the states probed and hence is readily related to the rotational temperature. A similar strategy, but using excitation from two separate vibrational levels, will be explored for monitoring the vibrational temperature. Initial results are encouraging, and experiments will continue on this topic during the coming year.

The second shock tube project involves development of PLIF diagnostics for studies of transverse jet injection into a supersonic crossflow. This flow is generic to the problem of fuel mixing in air-breathing scramjet engines, and hence the successful development and demonstration of a PLIF diagnostic for such flows would be valuable to the propulsion research community. Our approach has been to install a pulsed valve in the shock tube side wall which can be rapidly opened to provide a controlled flow of gas into the supersonic
crossflow produced by an incident shock wave. The use of shock wave heating allows a simple means of producing essentially any desired values of temperature and pressure in a supersonic crossflow. (Other facilities being developed for basic studies of jet mixing and combustion are much more limited, especially those which are intended for continuous operation. Furthermore, the continuous flow facilities are extremely limited in size, to the point that boundary layer effects may be significant, while the shock tube approach easily permits larger flow cross-sections.) Thus far we have studied two cases: (1) injection of NO into Ar/N₂ mixtures, to facilitate study of nonreacting flows; and (2) injection of H₂ into O₂/N₂ mixtures, with detection of OH, to study the combusting flow case.

An illustration of the flow studied and sample comparisons of reacting and nonreacting flow image data are given in Fig. 14. The upper panel of the color print is for the nonreacting case in which NO is imaged. Two images are shown, one acquired with a side-view, i.e. with the illumination in a central plane extending in the direction of the crossflow, and one acquired with an end-view, i.e., with the illumination transverse to the crossflow. The crossflow conditions for both flows are 2100 K, 0.4 atm and M = 1.4. Consider first the nonreacting flow results. Here the most important observation is that PLIF clearly reveals the presence of large scale structures in the flow. Also visible is the barrel shock and Mach disc structure commonly observed in supersonic free jets. Comparisons of several images shows that this supersonic portion of the jet remains constant, and only the portion of the jet downstream of the Mach disc undergoes strong oscillations. This observation of large scale structures is important, since it provides insight into the dominant mixing mechanisms present in the flow. Current modelling of the transverse jet mixing problem is based on a time-averaged view of the jet and hence is not physically realistic, even if the models include variable factors which can force agreement between various measured and calculated measurements of the extent of mixing. We believe that PLIF offers the opportunity of acquiring unique flowfield data which could be used to develop improved mixing models for this flow.

The lower panels of PLIF data in Fig. 14 illustrate OH distributions obtained for the same two views in the case of H₂ injection into hot air. These data clearly confirm that combustion has occurred, and that hot OH may be found in three distinct regions: (1) in the side wall boundary layer downstream of the valve orifice where the flow velocities are lower than the free stream, providing increased residence time; (2) in the small recirculation zone seen adjacent to the wall just upstream of the jet; and (3) in the mixing region associated with the shear layer separating the main fuel jet and the supersonic air flow. Note particularly that large scale structure is still observed (in the side view) with reacting flow, but that the structure seems more regular in orientation and size that in the nonreacting case. Apparently the energy release present in the reacting case has a significant effect on the mixing layer.
PLIF of Supersonic (Scramjet) Mixing and Combustion

- Use of shock tube provides access to a wide range of flow conditions

- First single-shot images of both nonreacting and reacting cases

- NO marks jet penetration and mixing; OH marks reaction zones and burnt gases

- Single-shot images reveal large-scale structures not apparent in averaged images

- Results confirm utility of PLIF measurements for scramjet research

Figure 14. PLIF imaging of transverse injection into a supersonic flow.
structure; hence a proper model of transverse jet mixing must depend on energy release. Inspection of the end-view image for the reacting case is also revealing in that it provides an indication of the relatively narrow width of the region containing OH 30 mm downstream (x/D = 15) of the jet orifice. Note also the significant level of OH present in the side wall boundary layer at this location. Clearly the combustion occurring in the side wall boundary layer is a key feature which must be considered in a realistic model of this flowfield. Further details regarding this work may be found in the publications by McMillin, Lee and Palmer cited in Secs. 3.2 and 3.3.

The results obtained thus far have been useful in planning improvements in the PLIF diagnostic strategies. For example, we realize now that an independent temperature measurement may also be needed to properly interpret the mixing process. Thus our recent effort has been directed toward a new two-laser, two-camera strategy which will hopefully allow simultaneous imaging of both temperature (coupled to energy release and extent of mixing) and species (best indicator of mixture fraction and extent of combustion). In addition, we are incorporating some modifications to the shock tube, such as a small backward-facing step upstream of the jet, which will increase the similarity of our flow to those under study in other laboratories.

To summarize, the work at Stanford has provided the first demonstration of PLIF both in hypersonic shock tunnel flows and in transverse jet mixing with supersonic crossflow. The results obtained thus far have been useful both in illustrating the potential of PLIF to researchers concerned with scramjet development and in guiding our plans for future work in this area. We continue to believe that PLIF will provide the type of data which will facilitate research and testing of scramjet concepts and components.

2.5 Shock Tube Diagnostics for Combustion Kinetics

In addition to providing an attractive test environment for developing laser diagnostics for gasdynamics and propulsion-related studies, shock tubes are recognized as the facilities of choice for measurements of reaction rate coefficients at high temperatures. Thus there is a continuing need to develop improved laser-based diagnostics for sensitive and accurate detection of species which are important in combustion. This has been a continuing element of our diagnostics program in recent years, and there have been several notable achievements. In particular, this program has led to the first use of cw ring dye lasers for species detection in shock tubes; and the list of species detected has now grown to include OH, NH, CH, NH₂, CN, NCO, NO and CH₃. The development of these diagnostics has led, in several cases, to the first direct measurements of rate coefficients for key combustion reactions at high temperatures. In addition, our work has led to the development of a new experimental approach to chemical kinetics in which a conventional shock tube is combined
with UV laser photolysis to allow direct study of reactions involving radical species at high temperatures. This experimental strategy has enabled study of reactions which were previously inaccessible to shock tube methods.

During the past year, our primary accomplishment has involved the establishment of a laser absorption technique for \( \text{CH}_3 \), a key radical in hydrocarbon combustion for which no previous shock tube technique was available. This work, still in progress, has been described in detail in the technical report by Davidson et al. listed in Sec. 3.3. Other key accomplishments of note have involved application of previously developed diagnostic methods to measure reaction rate coefficients of several reactions: \( \text{O} + \text{HNCO} \rightarrow \text{NH} + \text{CO}_2 \) and \( \text{O} + \text{HNCO} \rightarrow \text{OH} + \text{NCO} \); \( \text{C} + \text{H}_2 \rightarrow \text{CH} + \text{H} \) and \( \text{C} + \text{O}_2 \rightarrow \text{CO} + \text{O} \); \( \text{C} + \text{NO} \rightarrow \text{CN} + \text{O} \), \( \text{C} + \text{NO} \rightarrow \text{CO} + \text{N} \), and \( \text{CH} + \text{NO} \rightarrow \text{HCN} + \text{O} \); \( \text{CN} + \text{O} \rightarrow \text{CO} + \text{N} \) and \( \text{CN} + \text{O}_2 \rightarrow \text{NCO} + \text{O} \); and \( \text{H} + \text{HNCO} \rightarrow \text{NH}_2 + \text{CO} \). In addition, a detailed chemical model was developed for pyrolysis of methane dilute in an inert gas carrier. Details of these studies may be found in the papers listed in Secs. 3.2 and 3.3.

2.6 Digital Camera for High-Speed Imaging

Advances in diagnostic techniques for turbulent combustion are tightly coupled to state-of-the-art improvements in high-speed camera systems. For this reason, a portion of our overall program is directed at building a new camera with the capability for light-efficient, very high-speed recording of planar flow visualizations. In concert with this effort, we are developing experimental techniques for: (1) 2-d imaging of ultrafast combustion, plasma, and laser events, and (2) "instantaneous" 3-d imaging of turbulent flames and flows. During the past two years, we have assembled and empirically characterized a new high-speed camera system, and recently we began planar imaging experiments.

The camera system assembled at Stanford is shown in Figure 15. The Hadland image converter provides the fast framing capability, allowing recording at up to 50 million frames per second. Images are displayed on a phosphor screen in a flexible row-by-row format containing eight to 20 images per sequence. Recently Hadland, Hamamatsu and a group from Yale and Sandia began recording from image converters using lens-coupled CCDs. We have improved over such systems by using a tapered fiber-optic bundle to couple our image converter to an astronomy-grade CCD fitted with a fiber optic window. The unique fiber-optic train provides the spatial resolution
of a low-speed lens, yet is 2.5 to 10 times more light efficient than a fast f/2 lens. In short, the system offers high-resolution digital output with MHz framing capability and unmatched signal sensitivity.

Our first application of the camera system was conducted this year using the optical arrangement shown in Figure 16. Our imaging technique calls for planar illumination using a single 2-μs pulse from a flashlamp-pumped dye laser, the pulse duration being sufficient for up to 20 frames taken at 10 MHz. For 3-d imaging, a polygon mirror rotating at 30,000 RPM scans the laser sheet through the experiment while a stationary mirror adjacent to the scanner doubles the speed imparted to the scan sheet. For time-resolved sheet monitoring, we send a sample of the sheet onto a card in the camera's field of view, so a sheet profile appears with every frame. We also provide registration marks to assist post-processing of the images. The registration marks are created with fiber optic cables fed by a coordinated flashbulb. The resulting images are transferred to a PC for initial post-processing, then to our Pixar for volume rendering.

![Figure 16. Plan view of arrangement for 3-d imaging.](image)

Sample results are given in Figure 17 which shows soot scattering in a 6"-tall swirling ethylene diffusion flame along with the sheet monitor and registration marks. The 14-frame sequence was collected in 1.4 μs and hence represents instantaneous data. To assist interpretation, these same data have been volume rendered with our Pixar image processing system and the results are shown in Figure 18. Note the swirling character of the flame is only vaguely evident in the unrendered images, but is clearly visible in the twisted legs of the volume renderings. Note also the merits of 3-d information over 2-d by comparing any single frame of the unrendered data with the vastly more informative 3-d volume renderings of Figure 18.
Figure 17. Instantaneous 3-d visualization of a swirling ethylene diffusion flame (unrendered).

Figure 18. Instantaneous 3-d visualization of a swirling ethylene diffusion flame (rendered by Pixar).
In a second application, we have used the camera system to measure the 2-d transient characteristics of our dye laser. The circular patterns of Figure 19 show the evolution of the intensity distribution over the beam's cross-section measured at 100-ns intervals during a single laser pulse from our flashlamp-pumped dye laser. The results have been rendered by our Pixar to help illustrate the high-speed transients of the beam. This is believed the first time-resolved imaging of a dye laser, and the results clearly illustrate the need for time-resolved sheet monitoring in our 3-d experiments.

Figure 19. Evolution of a dye laser pulse.

In summary, progress has been steady in the development of this state-of-the-art high-speed digital camera system. We expect this unique system will enable many important extensions of planar imaging in combustion research.

2.7 High-Resolution PLIF Imaging in Turbulent Jets

One of the primary motivating issues for the development of planar laser-induced fluorescence imaging is the need for advanced diagnostics to study turbulence, and in particular turbulent combustion. The hope is that multi-dimensional data, of the type generated by PLIF, will provide improved insights into turbulence through the increased experimental information available. Since image data are quite different from the data gathered with traditional single-point observations, new questions have arisen in identifying the experimental quantities of greatest value and in selecting the types of image processing to be performed in reducing image data. In order to address these diagnostics questions, we have worked over the past several years to perform experiments in fundamental turbulent flowfields with the specific objectives of obtaining image data with: (1) high signal-to-noise
ratios; and (2) high spatial resolution. It was our belief that these two characteristics were necessary in order to guide work on image processing and in order for the work to have significant impact on the fluid mechanics community. The flowfield selected for primary study was a turbulent, nonreacting round jet at atmospheric conditions. The jet was nitrogen, seeded with biacetyl to a mole fraction of about 5%, flowing into a pure nitrogen background. Excitation of the biacetyl luminescence (primarily phosphorescence) was by a xenon fluoride excimer laser at 351 nm. This experimental strategy satisfied the requirement for high SNR, since biacetyl can be seeded at high levels and has a high conversion efficiency of absorbed-to-emitted photons. The high spatial resolution requirement was achieved through use a scientific-grade unintensified CCD camera. This was the first application of the now popular 384x576 Thomson array to flowfield imaging. Now, this same camera is in use in a large number of laboratories for basic and applied studies of fluid mechanics.

During the past year we have completed two tasks of note, discussed below: (1) development of user-friendly image processing software to aid interpretation of turbulence data; and (2) a survey of flowfield tracer materials with a short radiative lifetime needed to probe high-speed flows.

Our image processing is carried out on a Pixar computer, which is a relatively fast machine designed for the processing and interactive display of measured or computed image data. This machine is now four years old, and over this period we have built up unique capability for processing and display of PLIF data. A current example, illustrating work carried out this year, is shown in Fig. 20. Shown here, in the left panel, is a typical single-shot PLIF image of the jet mole fraction (concentration) in a transverse plane of a Re = 5000 nonreacting jet. The large-scale structures, now accepted as common to such flows, are apparent. This data set is manipulated in two ways in this figure. Firstly, the local gradient of the concentration is calculated (at each image location) and squared. This result, also in the left panel, is relevant since the square of the gradient is closely related to the scalar dissipation or mixing rate in turbulent mixing. As an alternative and complementary form of data display, the right-hand panel shows a landscape rendering of the jet concentration, with color coding of the local gradient. These two examples of image data manipulation illustrate the potential of computer processing to enhance human interpretation of flowfield data.

Our second activity in high resolution imaging has involved a survey of alternatives to the use of biacetyl as a flowfield tracer. In particular, we seek a tracer with a shorter radiative lifetime so that it can be utilized in high speed flows without "blurring" of the image. The biacetyl emission employed in our past work is known as phosphorescence, meaning that the radiation is relatively long-lived (about 1 millisecond at our conditions), although a smaller (about one part in 78) signal is also present owing to fluorescence. The fluorescence lifetime
IMPROVED RENDERING TECHNIQUE ENABLES VISUALIZATION OF MULTIPLE PARAMETERS

\[ \text{N}_2 \text{ Jet (Re = 5000). Instantaneous tracer concentration and squared magnitudes of the gradient} \]

Concentration landscape rendering with color coded gradient information

- 2-D data are displayed by landscape rendering
- One parameter is mapped by height. Second parameter is encoded by color
- Shades, perspective and rotation increase depth perception
- Potential application to combined velocity-temperature displays (|\vec{v}| - T, |\vec{v}||\nabla T|)

Figure 20. New capabilities for computer processing and display of PLIF data.

Hanson / Stanford
in biacetyl is much shorter, about 50 nsec, and hence the fluorescence emission following a short laser excitation pulse (typically 20 nsec) is complete before the illuminated gas sample has moved a significant distance. Unfortunately, the weakness of biacetyl fluorescence inhibits its use. An ideal tracer, suitable for applications in high speed flows, would have a high vapor pressure (so that it can be seeded at high mole fractions), a high fluorescence quantum efficiency (i.e., conversion of absorbed photons to fluorescence photons), and compatibility with existing pulsed laser sources. After an extensive literature survey, and a multi-month laboratory effort, we have selected acetone as the preferred "fluorescence tracer". A paper summarizing this activity is now in preparation, but we can summarize the primary characteristics of acetone here: (1) acetone has high vapor pressure, about 180 torr at room temperature, so that it can be easily seeded at high levels; (2) it is economical, easily handled and non-toxic, so that safety is not a problem; (3) the peak absorption is at 275 nm, but reasonably efficient excitation occurs at the convenient laser wavelengths of 248 nm, 266 nm and 308 nm; (4) the fluorescence quantum yield is about 0.2%; (5) the emission has a short lifetime (4 nsec) and occurs in a wide band which peaks at 480 nm, in the visible, so that it can be recorded on an unintensified camera; and (6) we estimate that, at room temperature and with existing laser sources, the fluorescence signals achievable with acetone are only about a factor of six less than the high phosphorescence signals obtained with biacetyl. Thus, we believe that it will be possible to obtain both high SNR and high spatial resolution, comparable to our past experience with biacetyl, in high speed flows seeded with acetone. Experimental work to verify this new approach will be carried out during the forthcoming year.

### 2.8 Multiplex Degenerate Four-Wave Mixing

During the past year we initiated work to explore diagnostics concepts based on degenerate four-wave mixing (DFWM). DFWM is a nonlinear optical process, with similarities to CARS, which has been under study in the nonlinear optics community for about a decade, owing largely to the phase conjugate nature of the signal beam generated. More recently, DFWM has been adopted by the combustion diagnostics community as a promising new approach which may offer advantages over CARS and LIF in some cases. Owing to the complexity of the physics, however, progress has been relatively slow in developing a quantitative understanding of the relevant processes, but there have been a handful of impressive demonstrations which suggest that useful signal levels can be achieved. Our goal has been to explore DFWM as a possible means of circumventing the two primary limitations of PLIF: (1) the difficulty of applying PLIF in highly luminous flows, particularly if the flowfield is large; and (2) the inability of PLIF to access all species. We are particularly interested in DFWM strategies for plasma diagnostics, since our current effort to apply LIF to atmospheric pressure plasmas suggests that luminosity will restrict LIF
diagnostics to below about 8000 K. Owing to the fact that the signal beam generated in the four-wave mixing process is highly collimated, it is possible to locate the detection optics far from the experiment and thereby to reduce the background signal due to luminosity to acceptable levels, even for plasmas in excess of 8000 K.

We have been pursuing a variation of DFWM known as multiplex DFWM in which the laser source has sufficient spectral width to excite multiple transitions simultaneously. As a vehicle to understand the relevant physics, we've applied the concept to excite two adjacent rovibrational lines of OH in a simple flame. The pulsed dye laser has been modified so that it produces a well-behaved, 1 cm$^{-1}$ bandwidth pulse near 311 nm, thereby exciting the $P_1(8)$ and $Q_2(14)$ transitions of the (0,0) band of the A$\rightarrow$X system of OH. The experimental arrangement is shown in Fig. 21. This is known as the phase-conjugation geometry; here two strong counterpropagating, collimated pump beams are overlapped in the interaction region by a weak probe beam. The resonant nonlinear interaction of the three beams with the molecules in the probe volume generates a fourth conjugate signal beam of the same frequency, which is also collimated and propagates backward along the path of the probe beam. The strength of the signal beam is a complex function of several parameters, but generally is proportional to the square of the number density of molecules in the participating absorption states. The essence of the multiplex concept is to simultaneously excite two transitions and to use the measured ratio of signal intensities at the wavelengths corresponding to the two lines to infer the relative populations and hence temperature. An etalon is used to provide a spectral distribution of the signal beam; this is recorded on a CCD array to allow either single-shot or frame-averaged recording.

A sample result of our first experimental result is shown in Fig. 22. Here the intensity distribution in the signal beam is shown; the two features are the two absorption transitions excited and the ratio of the two signal peaks can be converted to temperature. Good agreement is found with thermocouple measurements in a H$_2$/O$_2$ flame. Details will be provided in a publication now being prepared by Dr. Yip who conceived and conducted the experimental strategy. Although the multiplex DFWM concept was first advanced by another group (Ewart, 1989, Optics Letters), we believe that this is the first practical application of these ideas to measure temperature in gas flows.

Work is now being planned to explore another variation of DFWM which may allow provide the basis of a velocity diagnostic in low pressure plasmas. That effort will continue during the next year.
Figure 21. Experimental arrangement used for multiplex degenerate four-wave mixing thermometry.

Figure 22. Multiplex DFWM spectrum obtained using a single-laser-shot. Two peaks corresponding to DFWM resonant with the P1(8) and Q2(14) transitions of the (0,0) band of the A→X system of OH are clearly visible.
3.0 PRESENTATIONS AND PUBLICATIONS

3.1 Presentations (10/90 – 10/91)


3.2 Refereed Publications (10/90 – 10/91)


3.3 Technical Reports (10/90 – 10/91)


4.0 PERSONNEL

Individual researchers supported by the program are listed below. All the work has been carried out in the High Temperature Gasdynamic Laboratory, in the Department of Mechanical Engineering, under the supervision of Professor R. K. Hanson.

4.1 Postdoctoral Research Associates

Dr. B. Yip (50% time)
Dr. D. F. Davidson (20% time)
Dr. P. Arroyo (100% time)
Dr. J. Seitzman (25% time)

4.2 Graduate Research Assistants

Jerry Seitzman
Mike Lee
Doug Baer
Brian McMillin
Dave Hofeldt
Antonio Lozano
Paul Danehy
Brian Patrie
Hwa-Jin Chang

4.3 Ph.D. Degrees Awarded

Dr. Jerry Seitzman, 6/91, "Quantitative Applications of Fluorescence Imaging in Combustion"

Dr. David Hofeldt, 11/90, "Instantaneous Imaging Diagnostics for Measuring Particle Sizes and Spatial Distributions over Extended Regions in Two-Phase Flows"

Dr. Larry Cohen, 11/90, "Emission and Laser-Induced Fluorescence Diagnostics of a Supersonic Jet of Plasma-Heated Nitrogen"
5.0 SIGNIFICANT INTERACTIONS

In addition to the interactions associated with the presentations and publications listed in Section 3, we have had numerous visitors to our laboratory during this past year. Foreign visitors have come from Germany, France, Holland, Great Britain, Canada, Spain, Portugal and Japan; industrial and national laboratory visitors have included representatives from Rocketdyne, Aerometrics, Physical Sciences, Lockheed, Boeing, Metrolaser, AEDC, NASA Ames, NASA Lewis, NIST, Sandia, Lawrence Livermore, General Motors and Ford. Professor Hanson has given invited presentations on AFOSR-sponsored diagnostics research to industrial laboratories and government groups in the U.S., Europe and Japan. Members of our group have provided technical information and advice, by telephone and mail, to several external researchers interested in duplicating or extending our diagnostics concepts.

Interest in the potential application of advanced laser diagnostics to various practical problems, especially associated with hypersonic flow and the NASP program, continues at a high level, and the AFOSR-sponsored program at Stanford has achieved a high level of recognition for its contributions to this field. The increased interest we are witnessing is a useful indicator of the extent of technology transfer which is occurring in laser diagnostics between Stanford and industrial and government labs. A sustained research effort, at Stanford and other university labs active in diagnostics research, is required, however, to ensure the success of this technology transfer.