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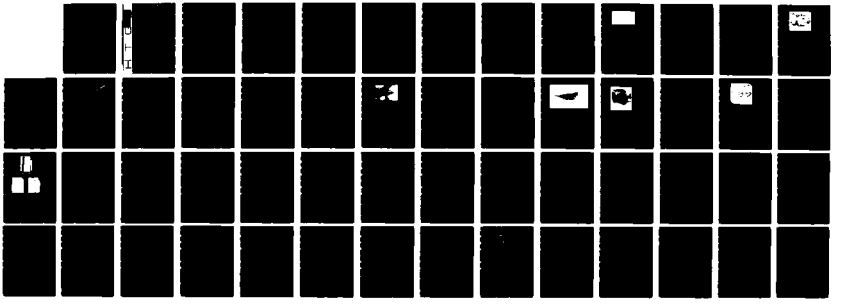

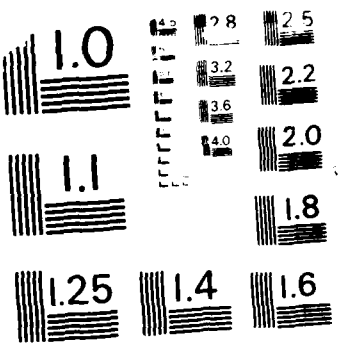

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Annual Scientific Report

on

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Progress is reported for the past year of an interdisciplinary program to establish advanced optical diagnostic techniques applicable to combustion and plasma flows. The primary effort is on digital flowfield imaging techniques, which offer significant potential for a wide range of spatially resolved 2-d and 3-d measurements. The imaging is accomplished by recording light scattered from a planar laser-illuminated region using a modern solid-state camera. The scattering process is generally laser-induced fluorescence, though Mie scattering is also used in connection with sizing particles. Activities reported herein include: (1) single-photon fluorescence imaging of molecular oxygen; (2) two-photon fluorescence imaging of CO and H_2O; (3) photofragmentation-based fluorescence imaging; (4) multiple particle sizing by imaging of Mie-scattered light; (5) fluorescence-based velocity and pressure imaging; (6) 3-d imaging; (7) laser and solid-state camera development; and (8) processing/display of image data using a specialized image computer. Other diagnostics topics discussed in this report include research on laser wavelength modulation spectroscopy and development of plasma diagnostics based on laser-induced fluorescence and Stark broadening. Finally, initial work to develop a laser photolysis shock tube, for fundamental studies of chemical kinetics and spectroscopy in high temperature gases, is described.

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TABLE OF CONTENTS

1.0 INTRODUCTION

Progress is reported for the past year of an interdisciplinary program to establish advanced optical diagnostic techniques applicable to combustion and plasma flows. The primary effort is on digital flowfield imaging techniques, which offer significant potential for a wide range of spatially resolved 2-d and 3-d measurements. The imaging is accomplished by recording light scattered from a planar laser-illuminated region using a modern solid-state camera. The scattering process is generally laser-induced fluorescence, though Mie scattering is also used in connection with sizing particles. Imaging-related activities reported herein include: (1) single-photon fluorescence imaging of molecular oxygen; (2) two-photon fluorescence imaging of CO and H_2O; (3) photofragmentation-based fluorescence imaging; (4) multiple particle sizing by imaging of Mie-scattered light; (5) fluorescence-based velocity and pressure imaging; (6) 3-d imaging; (7) laser and solid-state camera development; and (8) processing/display of image data using a specialized image computer. Other diagnostics topics discussed in this report include research on laser wavelength modulation spectroscopy and development of plasma diagnostics based on laser-induced fluorescence and Stark broadening. Finally, initial work to develop a laser photolysis shock tube, for fundamental studies of chemical kinetics and spectroscopy in high temperature gases, is described.

2.0 PROJECT SUMMARIES

Included in this section are summaries of progress in each of four project areas. In most cases each project summary contains the following subsections: (a) Introduction; (b) Scientific Merit; (c) Status Report; (d) Publications and Presentations; (e) Personnel. Additional descriptions of this work may be found in the cited publications and in previous annual scientific reports.

2.1 Digital Flowfield Imaging

Introduction

The utility of flow visualization as a diagnostic in studies of fluid mechanics and combustion is well established. Until recently, however, most visualization techniques have been qualitative and based on line-of-sight approaches which lack species specificity and are poorly suited for flows with three-dimensional characteristics. With the recent development of laser-based light scattering techniques, it has become possible to

obtain spatially and temporally resolved quantitative records of flow properties throughout a plane using sheet illumination and a scattering technique such as Raman, fluorescence or Mie scattering. Such multipoint measurements essentially provide "images" of the flowfield property being monitored; as these images can be recorded on modern solid-state array detectors, coupled to a computer for analysis and display, we call this approach "Digital Flowfield Imaging." Work along these lines is in progress at Stanford, Yale, SRI, Sandia (Livermore) and the Aeropropulsion Lab at Wright Field.

As examples of the capability of these new methods, in our laboratory at Stanford we have made instantaneous (10 nsec), multiple-point measurements of several species (OH, NO, O_2, CH, C_2, C_2H_2, CO, Na and biacetyl) in a variety of flames and nonreacting flows using planar laser-induced fluorescence (PLIF), and we have initiated work to provide simultaneous particle size and spacing measurements in spray flames using planar Mie scattering (PMS). Initially we employed a 100x100 element (10^4 pixels) camera, but more recently we have begun to utilize larger detector arrays with up to 384x576 elements (>220,000 pixels!) and have plans to test a megapixel camera. The sensitivity demonstrated thus far for molecular species is in the 10s of ppm range, with spatial resolution typically much better than 1 mm. Of equal importance, we have recently demonstrated two variations of PLIF which yield temperature, and another variation which enables simultaneous measurements of pressure and velocity (see Section 2.1(h)) without particle seeding. These new techniques provide significant advances in measurement capability with potential scientific impact extending well beyond the field of combustion for which the methods were originally developed. Finally, we note: (1) that these imaging methods have been extended to 3-d, by rapid scanning of the illumination plane; and (2) the quantity and quality of image data which can now be acquired has stimulated research on processing and display of image data using a recently acquired Pixar computer.

Scientific Merit

Digital flowfield imaging has the potential to stimulate scientific advances in several fields including fluid mechanics, combustion and plasma sciences, and supersonic aerodynamics. Our research also contributes to the advancement of related technologies, such as lasers and image processing, and it adds to the fundamental data bases for spectroscopy and reaction kinetics of high temperature gases and plasmas. The Stanford program has made pioneering contributions to flowfield imaging, particularly with regard to the establishment of new strategies for sensing the several flowfield parameters of

interest, and to the implementation of intensified camera systems which incorporate recent advances in intensifiers and array detectors interfaced with laboratory microcomputers.

Status Report

Our work on flowfield imaging has been detailed in a series of papers and reports (see list at end of this section), and so here we focus only on recent activities. For convenience and clarity we provide separate status reports on: Oxygen Imaging, Two-Photon-Excitation Fluorescence, Photofragmentation Excitation, Multiple Particle Sizing by Imaging, System Improvements, Image Processing Computer, 3-d Imaging, and Velocity and Pressure Imaging.

(a) Oxygen Imaging

Previously we have applied single-photon-excitation PLIF to visualize OH, Na, NO, C_2, CH, biacetyl and I_2 in several flows. Recently we have dedicated a significant part of our overall effort to developing measurement capability for O_2, as we discuss briefly below. Molecular oxygen is of particular interest in combustion, aerodynamics and heat transfer studies, but is usually considered to be inaccessible by optical techniques as room temperature O_2 absorbs strongly only in the UV below 200 nm. In addition, fluorescence is known to be weak owing to strong predissociation effects. Two factors mitigate this conventional viewpoint: (1) the recent development of high energy, tunable UV and VUV laser sources, particularly excimer and Raman-shifted excimer lasers; and (2) the fact that the dominant absorption transitions at high temperatures are very much stronger than the (separate) transitions normally employed for monitoring O_2 at atmospheric conditions.

Following an initial literature survey of relevant past work on O_2, we have maintained a continuing effort to develop and refine a set of computer codes which allow prediction of O_2 absorption and fluorescence spectra as a function of temperature, excitation wavelength, laser spectral bandwidth, and spectral filtering in the fluorescence collection channel (see, for example, paper 23 in list at the end of this section for published results using our first computer model). The calculations are complex and will not be described here, but in summary we find that there are several attractive choices for the laser source to be used in PLIF imaging of O_2. The simplest choice is to use a conventional broadband ArF laser ($\Delta\nu$ = 200 cm^{-1} at 193 nm), which is well suited for high temperature

applications, or Raman-shifted ArF (179 nm for H_2 and 183 nm for D_2) for low temperature applications. Such lasers are readily available and have high pulse energy which leads to good signal-to-noise ratios in the PLIF signals. In flows which are either reactive (varying O_2 mole fraction) or compressible (varying density), however, it is necessary to make multiple measurements to separate the signal dependence on temperature (Boltzmann fraction in absorbing state), total mass density and O_2 mole fraction. In these cases, which apply to both combusting flows and supersonic flows, there are several possible strategies involving multiple excitation wavelengths and multiple spectral collection channels. For example, one might utilize both the fundamental (unshifted) and Raman-shifted light from a single broadband excimer laser pulse, with one beam delayed before passage through the measurement region, and two broadband-collection cameras to infer both temperature and O_2 concentration in a combustion flow. Alternatively, two lasers, for example one tunable narrowband excimer and one broadband excimer laser, and two cameras might be used to generate two fluorescence signals at each flowfield location. The optimum strategy depends on the problem under investigation and the equipment available.

Until recently, only broadband excimer lasers were available, and our initial measurements were carried out with such a system (Lambda-Physik EMG 203, operating on ArF at 193 nm). O_2 images were obtained in a number of flames and reported in paper 21 (listed at end of section). These were the first 2-d images of O_2 obtained in any flowfield and were regarded as a significant development owing to the broad importance of O_2 in many engineering fields. Subsequently, we used the same laser to perform PLIF imaging of temperature in heated air jets (see paper 26 for details); here the mole fraction of O_2 was constant and hence the PLIF signal varied only with temperature.

During the past year we have acquired a tunable, narrowband (0.3 to 1.0 cm^{-1}) excimer laser (Lambda-Physik EMG 160), and we have begun to investigate PLIF imaging of O_2 using this new laser source. The virtue of this laser is that all (or nearly all, depending on absorption linewidth) of the pulse energy can be used to excite specific absorption transitions. This is a more efficient use of the laser energy (little energy is at nonabsorbing wavelengths) and, in addition, allows selection of transitions with varying temperature dependence. Although work remains to fully characterize the laser performance, and to make minor modifications specific to our needs, we expect that this laser will prove to be the optimum choice for O_2 imaging. At present we are setting up experiments with this new laser to validate our O_2 spectroscopy model and to explore candidate multiple-wavelength strategies for simultaneous determination of O_2

concentration and temperature in reacting, isobaric flows. Exploration of schemes to deal with reacting compressible (supersonic) flows is also planned.

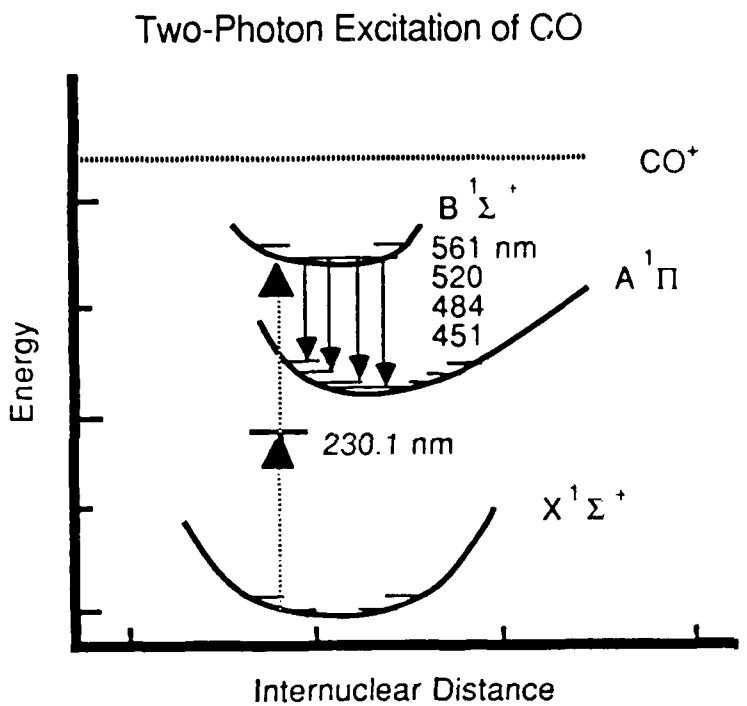
(b) Two-Photon-Excitation Fluorescence

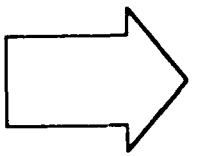
The work described in the previous section was based on single-photon excitation, that is the fluorescent emission is preceded by absorption of single photons which raise the molecule in one step from a discrete lower energy level to a discrete upper level. Recently, we have begun to explore two-photon absorption as a means of inducing fluorescence in stable molecules which have no strong, accessible one-photon absorption spectra. Most of our effort has been directed toward CO, a species which is of interest both because it is a stable intermediate in combustion and because it can serve as a relatively simple fuel in fundamental studies. A schematic of the basic concept and illustrative imaging results for the instantaneous CO distribution in a laminar diffusion flame (CO in air) are presented in Fig. 1. The work is now complete and details, including successful measurements in cold CO jets, CO diffusion flames, and premixed CH_4-air flames, are given in two publications (papers 30 and 35).

The primary virtue of two-photon-excitation fluorescence is spectroscopic access to additional molecular species. There are at least two significant liabilities to this approach, however, namely that the process is nonlinear and it is generally weak. The nonlinearity issue can complicate calibration procedures to convert fluorescence data to absolute CO concentrations. The weakness of the process leads to use of high pulse energies which can perturb the system being studied by various mechanisms. Our experience with CO was quite favorable in that the signal levels obtained (with only 2 mJ/pulse at 230.1 nm) were quite satisfactory for imaging, and a relatively simple physical model accounting for energy transfer and photoionization processes was sufficient to model the intensity dependence of the observed signals. It was necessary, however, to employ a multipass cell in these experiments to achieve single-shot imaging.

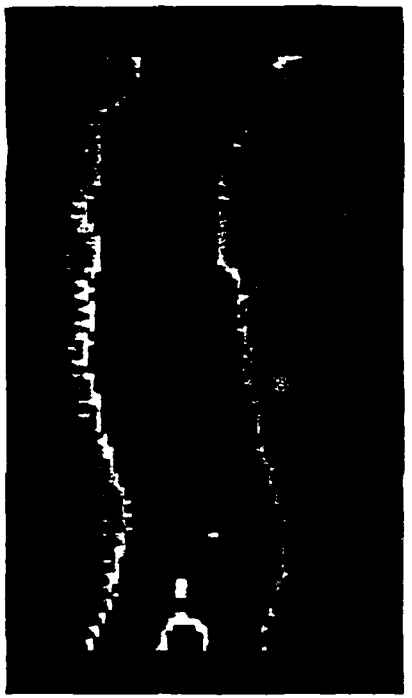
Very recently we have attempted two-photon-excitation experiments with H_2O, which is another critical combustion species that has not yet yielded to conventional single-photon imaging. The strategy pursued was to excite with a krypton fluoride (KrF) excimer laser at 248 nm which is known, from experiments at low pressures, to absorb via a coherent two-photon process with subsequent rapid dissociation to OH + H. In past work, conducted in other laboratories, it was found that the OH was produced primarily in an excited electronic state ($A^2\Sigma$) and hence subsequently emitted fluorescent light corresponding to the $A^2\Sigma \rightarrow X^2\Pi$ transition (near 300 nm). Thus the overall process might be termed photofragmentation fluorescence, and the emitted light, although due to

NEW TECHNIQUE ALLOWS QUANTITATIVE MEASUREMENT OF PREVIOUSLY UNDETECTABLE SPECIES





Array Camera Detects Visible Fluorescence



Instantaneous CO Distribution In Laminar Diffusion Flame

- First 2-D Images Based on 2-Photon Excitation
- New Photoionization Strategy Renders Fluorescence Quantitative
- Potential Applications to Other Stable Species

Figure 1. Two-photon excitation fluorescence imaging of CO.

OH decay, can be related to the original H_2O concentration. Unfortunately, in the experiments conducted in our laboratory at atmospheric pressure, and for a variety of H_2O-containing systems at elevated temperatures, no significant emission was observed at the expected wavelengths. We attribute our failure to observe OH fluorescence to pressure-dependent changes in the several competing dynamical processes which occur subsequent to the initial two-photon excitation in H_2O. Our current effort to image H_2O is directed toward use of single-photon photofragmentation fluorescence using 193 nm laser light, as discussed in the following section.

(c) Photofragmentation Excitation

As indicated above in subsection (b), many critical combustion species cannot be imaged by conventional single-photon-excitation fluorescence. Alternative fluorescence schemes include two-photon excitation (subsection (b) above) and photofragmentation. In photofragmentation, absorption of one or more photons is used to populate an electronically excited state of the species being monitored. If this is a dissociative state, or if the molecule undergoes rapid transfer to a dissociative state prior to radiative decay, then fragment molecules (i.e., the "photofragments") may be formed in excited states which subsequently emit by spontaneous emission. This fluorescence light, termed "photofragmentation fluorescence," may be imaged as a measure of the original absorbing species concentration. If the photofragments are formed in their ground electronic states, then a second laser may be used to perform fluorescence imaging of the fragments, but this scheme has the disadvantage of requiring two laser sources.

The utility of photofragmentation has been significantly enhanced by the development of excimer lasers, which provide intense pulses of UV and VUV radiation with sufficient energy to break even relatively stable chemical bonds. In the case of hydrocarbons, for example, many of the species of common interest have been found to be dissociated by single- or multi-photon processes using either ArF (193 nm) or KrF (248 nm) lasers. A difficulty of the method, in fact, is that in gaseous mixtures there may be a number of photofragmentation processes occurring simultaneously. This can greatly complicate the quantitative interpretation of the fluorescence signals observed.

The primary study of photofragmentation fluorescence in our laboratory has been in spray flames. Of particular interest has been the strong photodissociation of C_2H_2, via a two-photon process using 193 nm photons from an ArF laser, which leads to emission from CH* near 430 nm. We've employed this imaging scheme to reveal regions containing unburned fuel vapor, since acetylene is a known to be a primary constituent of hot fuel vapor for a broad range of liquid fuels. A schematic of the method and a sample

result are shown in Fig. 2. Further details of this work are given in paper 25. Although the data cannot readily be converted to quantitative images of C_2H_2 concentration, the data are quantitative in a spatial sense. That is, the shape and location of zones containing fuel vapor are accurately determined and may be used together with images of flame fronts and spray location for various spatial comparisons and correlations. This information alone is sufficient to address several fundamental questions regarding spray combustion.

We have also briefly explored the photofragmentation concept for imaging H_2O. In contrast with the work described in subsection (b) which employed two-photon excitation at 248 nm, we have investigated the use of single-photon excitation at 193 nm (ArF laser). Although this work is still in progress and our conclusions are tentative, it appears that useful levels of OH are produced by this scheme when the water vapor is sufficiently hot. This suggests that the absorption process occurs from hot bands ($v'' \geq 1$). The OH produced, however, appears primarily in its ground electronic state, and hence it has been necessary thus far to employ a second, probe laser sheet at 308 nm (tunable XeCl excimer) to image the OH via PLIF. The signal levels obtained have been suitable for single-shot imaging of H_2O in a range of atmospheric pressure flame environments, though the quantitative interpretation of such images is still incomplete.

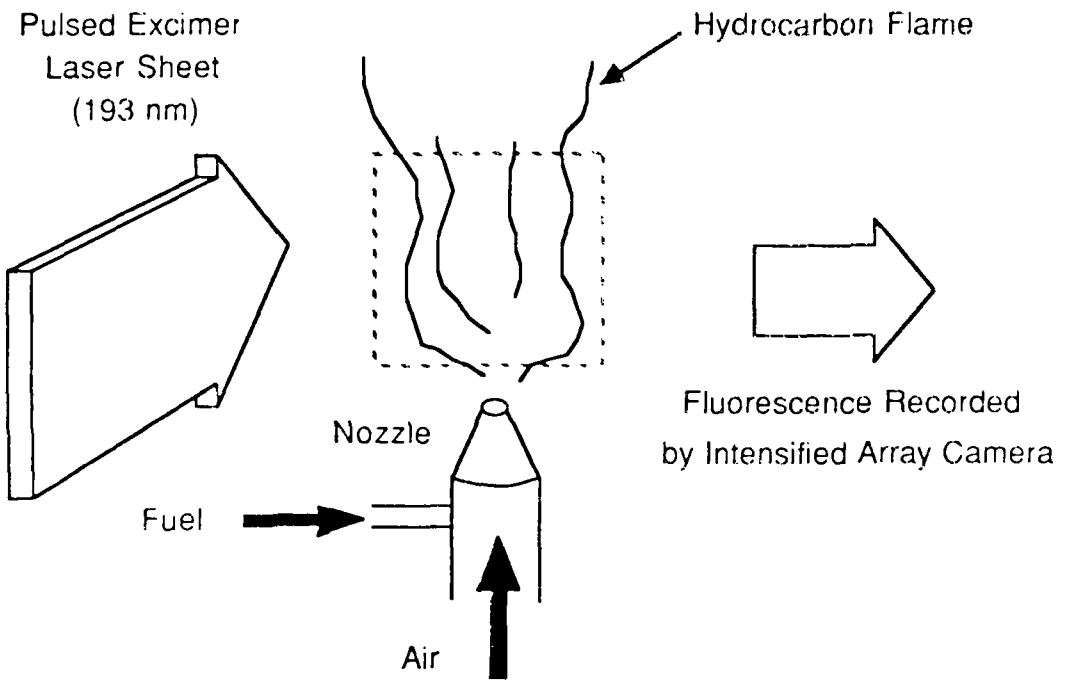
(d) Multiple Particle Sizing By Imaging

About two years ago we began to explore applications of our imaging system to particle sizing. The work was motivated by the considerable interest within the combustion community in sizing particles and droplets and by our observation that interference signals from Mie-scattered light frequently were a problem in our work imaging fluorescence in spray flames. These observations led to a new particle-sizing strategy which offers prospects of simultaneously sizing a large number of particles within a planar region. In brief, the region is illuminated by a sheet of broadband laser light, and the Mie-scattered light from particles in this region is imaged onto a solid-state array camera. In contrast with other schemes, however, the particles are sized by the amount of light scattered rather than by the size of the particle image. In most cases the size of the particle image is smaller than that of the recording camera element (pixel), and hence the large number of pixels available with modern detector arrays offers the prospect of sizing a large number of particles with each laser shot.

There are two critical aspects of this diagnostic concept which merit mention. First, to avoid the strong oscillations in scattering which result from surface resonances when monochromatic light is used, we have intentionally modified our dye laser to

PHOTODISSOCIATION CONCEPT ALLOWS IMAGING OF FUEL VAPOR

- Electronically Excited Fragments From Photodissociated Fuel Molecules Emit Light



Fuel Vapor Distribution
in Methanol-Air Spray Flame

- Reveals Spatial Structure in Fuel Decomposition
- Potential for Single-Shot Imaging of Fuel-Air Ratio

Figure 2. Photofragmentation scheme for fluorescence imaging of hydrocarbons

STANFORD/HANSON

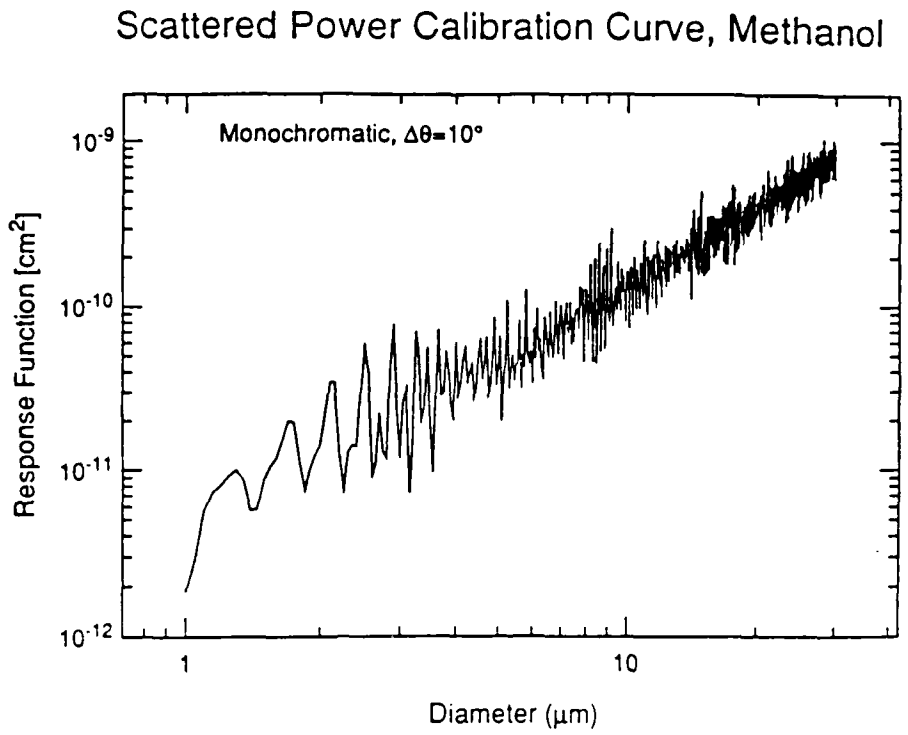
operate broadband. The spectral width achieved is about 15 nm (FWHM); the key modification is to degrade the dye laser oscillator to operate near lasing threshold. The result of using a broadband source is to smooth the usual resonance pattern, leading to pattern, leading to a monotonic increase in scattering cross-section with particle diameter, as is indicated in Fig. 3. For the conditions specific to our work the signal varies approximately with the particle diameter to the 1.8 power, and the resonances are smoothed sufficiently to allow use of this scheme for particle diameters greater than about 10 microns.

A second critical aspect of the method is that it requires uniform illumination intensity in the region where particles are being sized. Our plan for dealing with this constraint (see Fig. 4) is to utilize only the central portion of the illumination sheet, and to verify with a second, centrally located thin probe sheet that the scattering particles being measured are in the proper region. This approach requires two laser sheets and two cameras, all of which must be synchronized, but appears to work satisfactorily. This work is still in an early stage, but some details will be presented at two forthcoming meetings (see presentations 40 and 41 listed at end of section 2.1 and paper 36).

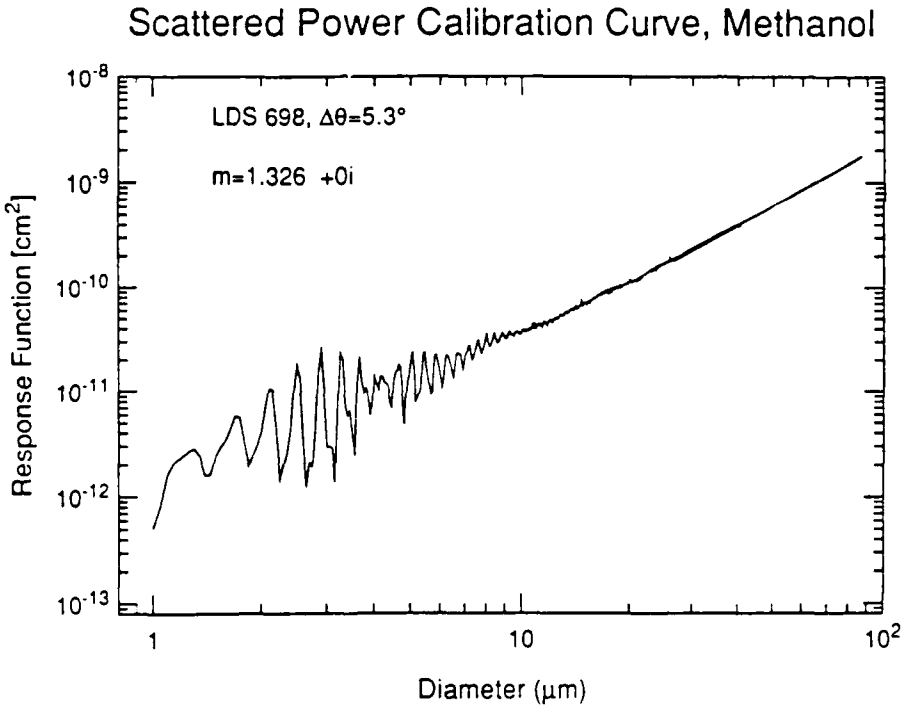
(e) Laser and Camera System Improvements

During the past year we have continued to devote a significant effort to the improvement of our imaging systems. In our early work on digital imaging we were forced to design and assemble a one-of-a-kind intensified camera and display system, using components drawn from a number of commercial sources and solving a number of interfacing and software problems on our own. This system was suitable for demonstrating feasibility of PLIF imaging in a variety of reacting flows, but it was not well suited for adoption by user-oriented researchers interested in acquiring similar capability. Moreover, rapid advances in laser sources and array sensor technology have created a situation where significant improvements in imaging system performance are now possible using more modern components.

For purposes of discussion, it is useful to consider PLIF imaging systems to be comprised of four major elements (see Fig. 5): the laser source, the experimental facility, the camera, and the image data readout/processing electronics. We focus here on improvements in laser sources and camera systems. Progress to upgrade our image data electronics through use of an "image computer" will be discussed in the next subsection.



(a)



(b)

Figure 3. Scattering response functions versus particle diameter for methanol droplets, m=1.326+09i, for circular apertures of specified width centered around $\theta=\phi=90^{\circ}$, x-polarized input beam. (a) Monochromatic, λ=695 nm; (b) Broadband dye laser spectrum using LDS 698 dye.

Experimental Setup

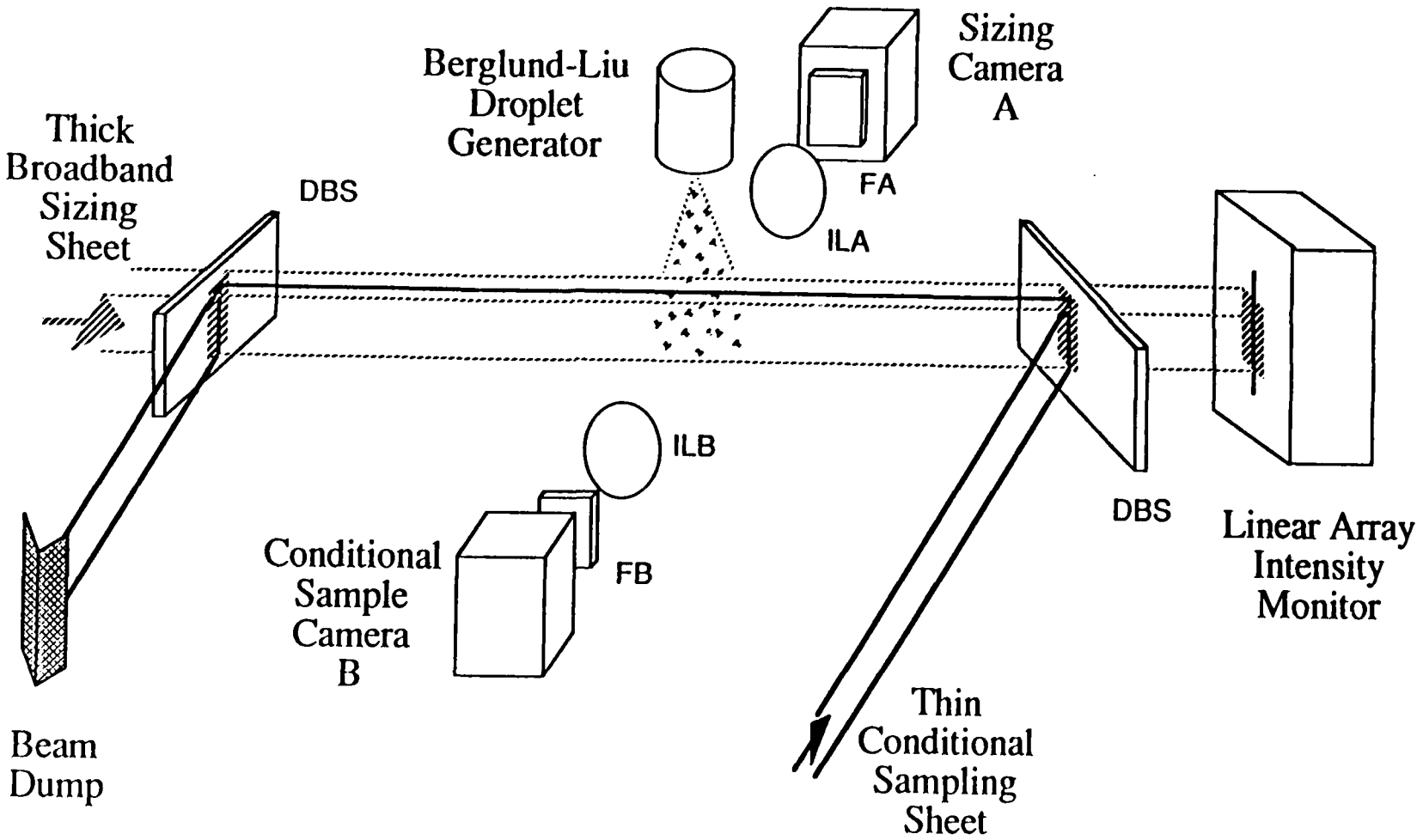


Figure 4. Experimental arrangement for particle sizing by imaging Mie scattered light.

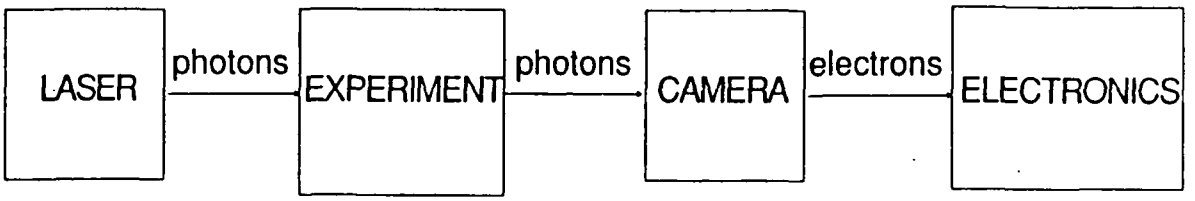


Figure 5. Elements of PLIF imaging system.

During the past year we have added two new laser systems to our laboratory: a tunable narrow-linewidth laser system (Lambda-Physik EMG 160) and a long-pulse-length flashlamp-pumped dye laser (Candela SLL 8000). The former laser arrived about a year ago and has already seen use in several experiments. Its main virtue is that the laser output is spectrally narrow (0.3 to 1.0 cm^{-1}) and can be tuned over the gain bandwidth (about 1 nm) at the primary output wavelengths of the laser, namely 193, 248, 308 and 351 nm. This important improvement over previous wideband excimer lasers allows much more efficient use of the laser energy, in terms of generating PLIF signals, and it also allows study of PLIF excitation schemes needed for simultaneously determining temperature and species concentration in reacting, compressible flows. This state-of-the-art laser (ours was the first of this model delivered in the U.S.) is a critical element in our research to image molecular oxygen in a variety of subsonic and supersonic flows. At present, we are carrying out minor modifications to this laser to facilitate measurements in supersonic air flows.

Our second new laser system is designed to provide high energy pulses of tunable laser radiation with a long (2 microsecond) pulselength. The utility of such a laser is twofold: (1) by utilizing long pulselengths at modest intensity levels, the integrated PLIF signals (in photons) can be greatly increased without saturating the transition probed; and (2) the long pulselengths provide time for recording a significant number (~20) of individual PLIF images. These individual images can be used to generate an "instantaneous" 3-d fluorescence image of the flow simply by rapidly sweeping the laser sheet location across the flowfield during the 2 microsecond period, or alternatively the sequential

images can be used to study a rapid transient in a fixed planar region. This laser system is currently undergoing installation in our laboratory and should be available for initial use within about 2 months.

As regards imaging camera systems, during the past year we have initiated work with two new systems and have continued developmental work on two other cameras which were acquired in previous years. Each of these systems has important attributes, and no one camera meets all the performance requirements of PLIF imaging experiments. It is helpful in discussing these various systems and their trade-offs to refer to the table of camera characteristics shown in Fig. 6. This table indicates that one major trade-off in camera selection is between framing speed and spatial resolution (number of pixels). Here we have subdivided available cameras into four major categories, with the highest spatial resolution camera appearing at the top and the highest framing rate camera appearing at the bottom of the table. All of these cameras can be utilized with or without intensification, though the difficulties involved in coupling an intensifier vary significantly.

CAMERA SYSTEM TRADE-OFFS

System	# Pixels	# Noise Electrons	Dynamic Range (# Bits)	Speed (FPS)	# Frames Stored (#F)	Notes
Hi Res	400 x 600 (4k x 4k)	< 5	14 - 16	0.1 (0.01)	50 / Disk (32 Mb each)	CCD Reformattable
Video	400 x 600	10^3	8	30 / 60	60 / RAM $>10^4$ /Tape	CCD Multiple Cameras Real-T Processing
Variable Framing	128 x 128	10^3	8	500	500 / RAM	PD / CCD Multiple Cameras Reformattable
Fast Framing	$200 \times \frac{1600}{\#F}$	< 5	8	10^3 - 10^8	8 - 32	Image Converter Coupled to Hi Res

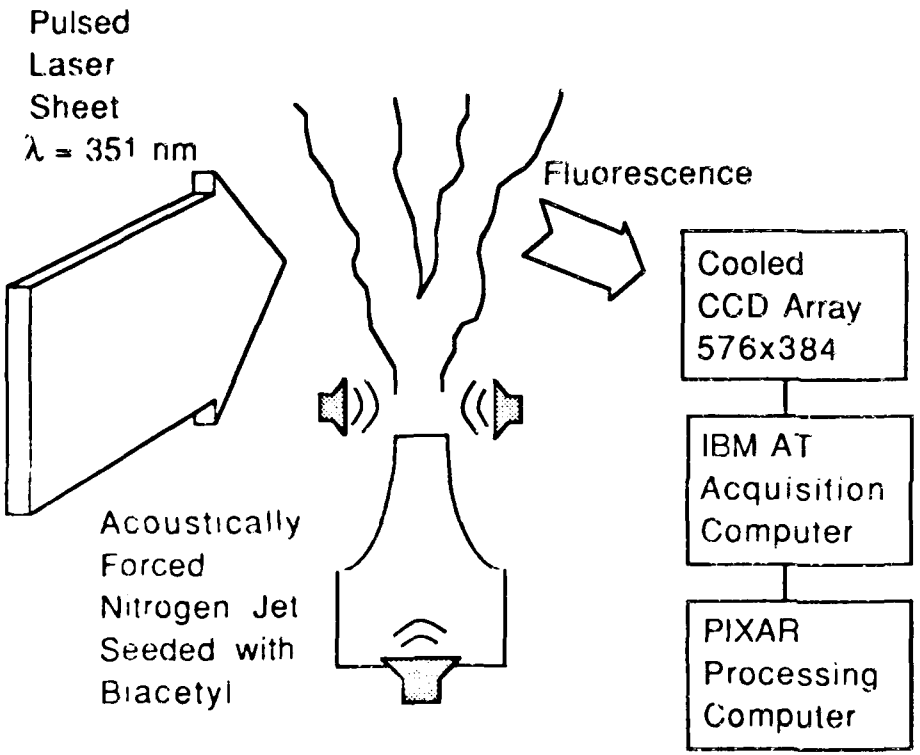
Figure 6. Table of imaging camera systems and their key characteristics.

In our earliest work, first reported in 1982 (see paper 1), we assembled a variable framing rate (up to 500 frames per second) 100x100 pixel camera based on photodiode array technology. That camera is currently being upgraded in our laboratory to a 128x128 configuration. The main virtues of this camera are its variable framing rate, paid for at the expense of relatively high noise level and modest spatial resolution, and its large pixel area. The latter property is advantageous when working with low light levels in order to obtain high values of SNR. At present the camera has been fiberoptically coupled to an intensifier and the system is operational. Work is in progress to couple the camera's dedicated computer (MicroVax I) to the laboratory Ethernet network so that image data sets can be transferred to the main laboratory computers (Vax 750, Sun 3 and Pixar Image Computer).

About three years ago, we began development of a high resolution camera (384 x 576 pixels) based on CCD technology. This camera provides a major increase in the number of pixels, to about 220,000 compared with 10,000 pixels in our original 100x100 camera. In addition, it has been designed to provide very low electron noise and hence large possible dynamic range. Our intent is that this astronomy-grade camera will be used for quantitative imaging applications where high spatial resolution and high values of SNR are needed. During the past year the camera has become operational and several image data sets have been acquired. An example PLIF image, shown in false color, for the instantaneous concentration of jet fluid in a bifurcating jet (biacetyl-laden N_2 into N_2) is shown in Fig. 7 (see also papers 28, 29 and 37). These data were acquired with a single excimer laser (XeF) pulse at 351 nm. At present the camera is functional though work continues to streamline its operation and make it more user-friendly. A remaining problem is that the camera is not intensified and hence is limited to use at visible wavelengths. Our work at Stanford with these large CCD arrays was among the first outside of the astronomy community, and we are convinced that the increased spatial resolution available will significantly impact the acceptance of PLIF imaging as a valuable research tool. While refining this camera system, we are tracking the availability of the next generation in high resolution sensors, namely 1k x 1k and 4k x 4k arrays with 1 and 16 megapixels respectively, which will have sufficient spatial resolution to enable direct observation of the complete range of turbulent flow structures (microscales to large-scale structures) in one camera frame.

Our major activity in system development at present is split between two new cameras, a "video" system and a fast-framing camera. The video system is intended to be a user-friendly, high-resolution camera compatible with the computer hardware and software which is rapidly becoming available for video imaging applications. Our goal is

TURBULENT FINE STRUCTURE DETAIL REVEALED BY LASER-INDUCED FLUORESCENCE



Mixing fraction in bifurcating jet

- Use of 221,184 Pixel Array Allows Fine Structure To Be Observed
- Factor of 5 Improvement in Spatial Resolution and >100 in Dynamic Range Over Previous Digital Imaging

Figure 7 Example application of high-resolution CCD in PLIF imaging of bifurcating gaseous jet

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to assemble a high performance camera, using readily available components, which can be copied by user-oriented researchers who wish to acquire PLIF imaging capability without a serious development commitment. Following a thorough survey, we selected an Amperex CCD array (NXA 1031/1061, 576x604 pixels) which has excellent noise characteristics and can be purchased with a bonded optical-fiber stub to enable intensification. (Most commercially available CCD arrays are not available with this critical feature.) This Amperex array operates on the frame-transfer principle, i.e., each field of the image is transferred by shift registers into a storage region during vertical blanking, and then clocked out serially to form the video signal during the subsequent field integration period. The array is read with a Data Translation frame-grabber board in an IBM AT computer. This frame-grabber has several useful image data operations available in hardware and software to facilitate a range of common image processing steps. At present we are assembling two camera systems, one intensified and one unintensified. Each system has two CCD arrays to allow dual image recording and is controlled by a single computer with two frame-grabber boards. The first system is now undergoing its first tests, and all the results obtained are very favorable. Once these two systems are completely checked out, we plan to make the design available to other interested researchers.

The last new system to be discussed is a fast-framing device based on an Imacon 790 image converter camera. The camera can be operated at several discrete framing rates; our system is set for 10 million FPS (frames per second). Up to 32 images can be recorded at this rate. Although the camera output is normally recorded with Polaroid film (which reads out the phosphor backplane of the Imacon), we intend to adapt a large CCD array to enable direct digital readout into computer memory. The camera will have two immediate applications: (1) 3-d imaging; and (2) rapid transient imaging. In both cases, the probable light source is the new Candela laser with its 2 microsecond pulse-length. At a framing rate of 10 million FPS, the laser source will allow 20 images to be recorded. For 3-d imaging the illumination sheet will be swept rapidly so that each image corresponds to a different illumination plane location. The camera has recently arrived and is currently undergoing installation in our laboratory.

(f) Image Processing Computer

The quantity of flowfield data associated with even a few individual PLIF images recorded on a high-resolution camera is quite impressive; such data sets rapidly enter the megapixel regime. Efficient handling and processing of such data quickly exceeds the capabilities of common lab computers. What is needed, in our opinion, is to utilize a

computer designed to deal with image data. Several such computers are now commercially available, and we anticipate a rapid growth in the use of such systems for "scientific imaging." Furthermore, it is likely that the common needs for processing images drawn from different branches of science will lead to rapid growth in software availability. In view of these trends we have acquired in the past few months a Pixar Image Computer which has been designed specifically for processing, interpretation and visualization (display) of large scientific data sets.

The key features of the Pixar are its large picture memory (48 megabytes), its high speed processor (40 MIPS), and its flexible control and programmability. Pixar is a relatively new company but is already recognized as a leader in image computing. Importantly, the company is located relatively close to Stanford, so that interaction with Pixar engineers and programmers is convenient. This has already been useful in bringing the system on-line.

We view the Pixar as a research tool, useful not only for routine processing of data but also for intelligent user interaction with data sets which may lead to new insights for fluid mechanics. For example, as 3-d data sets become available, and eventually 4-d (3-d plus time), there are key questions to be answered regarding the quantities to be extracted and displayed. A fast, user-friendly image computer should enable the researcher to explore data sets in creative ways and to seek new interpretations and models of fluid flow. In many ways, the evolution to 2-d, 3-d and 4-d data sets which is now occurring will put the experimentalists on par or ahead of computational fluid dynamicists (CFDers). The data sets which will be acquired will provide new challenges to flow modelers and should lead to more direct validation of evolving models of fluid flow.

An example Pixar computation is displayed in Fig. 8 for the case of the bifurcating jet data (see Fig. 7). Here the Pixar has converted the 2-d data (PLIF intensity in the central vertical plane of the jet) to a perspective view from a specific viewer orientation. Although only a single view is shown here, the Pixar enables scene rotation in real time on a high-resolution video monitor to give the viewer a sense of three dimensionality and to allow observation of details which can be seen from only a limited viewing angle.

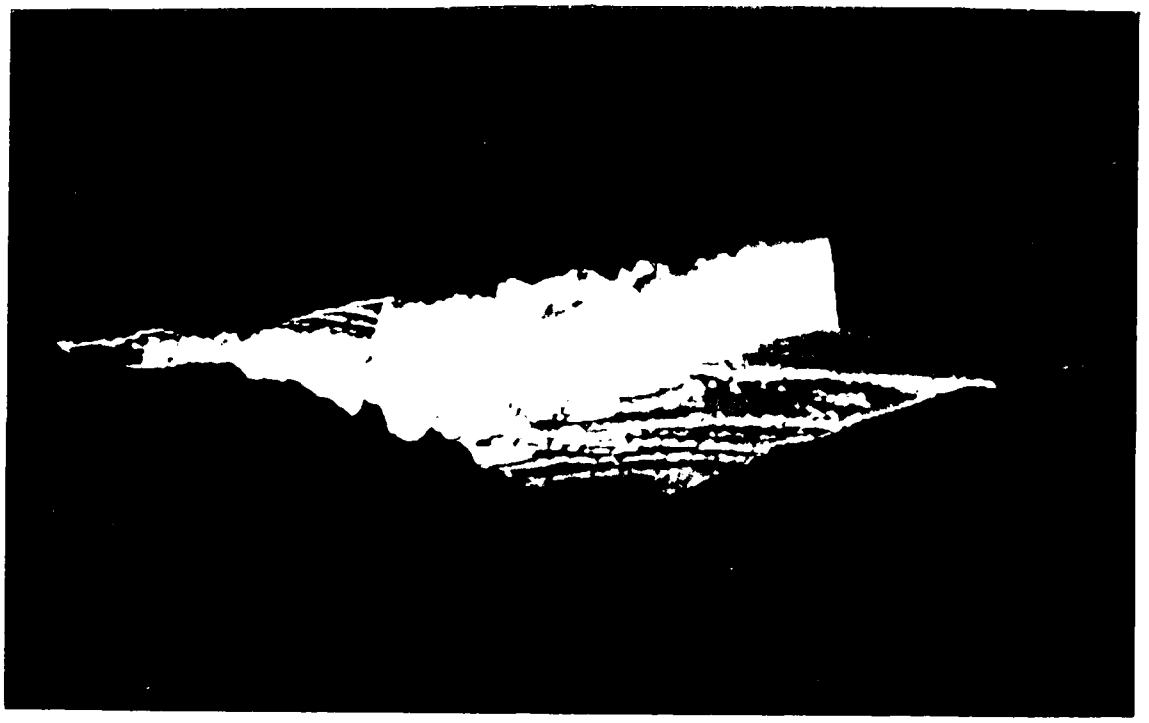


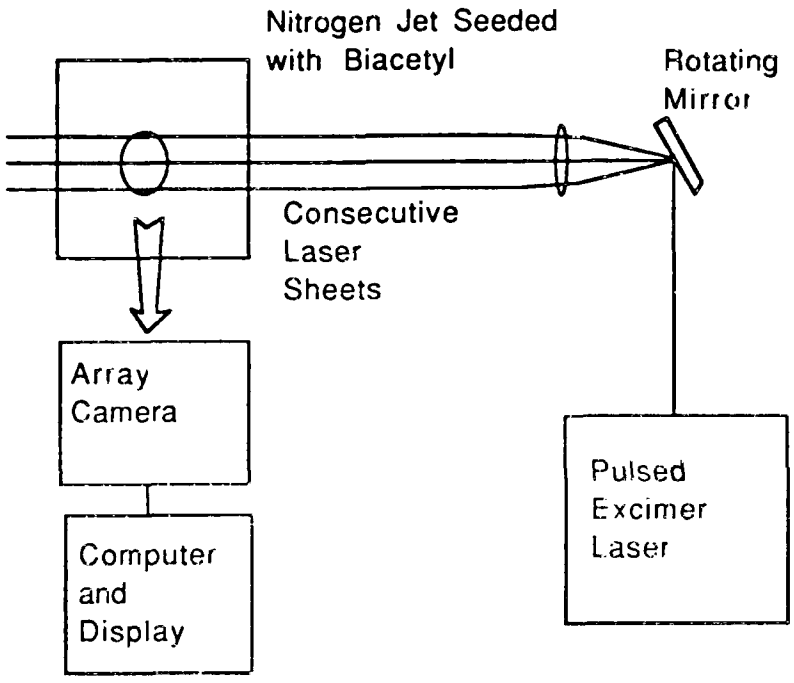
Figure 8. Perspective of view of mixture fraction (vertical axis) of bifurcating jet generated using PIXAR image computer. Flow from right to left.

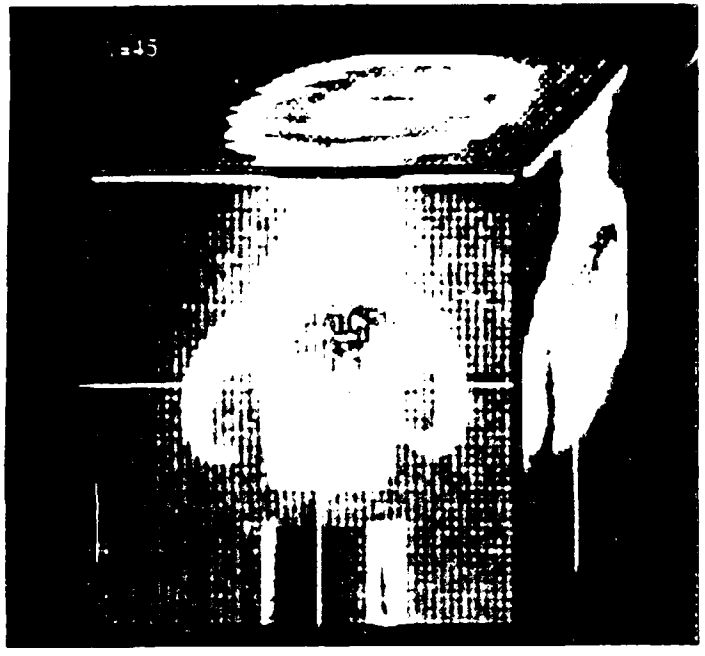
(g) 3-D Imaging

An important extension of our PLIF work during the past two years has been to record multiple planes of data, thereby generating the 3-d distribution of the imaged quantity. Thus far we have focussed on steady or repetitive flowfields, which permit slow scan rates for the illumination plane and the camera, but we will soon be able to record "instantaneous" 3-d distributions using the long pulselength Candela laser and the Imacon fast-framing camera. An example of our initial work on 3-d imaging, for the case of a biacetyl-seeded, axially forced jet, is displayed in Fig. 9 (details of this work are available in paper 37). The display used is known as a "cube display," in which the data stored along three orthogonal planes are shown on the three visible faces of the cube. The positions of the planes, indicated by the white cursor lines on the cube faces, are readily manipulated using our SUN workstation and software developed at Stanford. Rapid scanning of the displayed planes is also possible, providing the viewer with a sense of three dimensionality. Similar displays, but with greatly expanded data sets (up to 48 megapixels or voxels), will soon be possible using the Pixar computer.

Three Dimensional Digital Flowfield Imaging

- First 3-D fluorescence imaging in gases





Cube Display of 3-D Data
(240,000 pixels)

- Large data sets (up to 20 megapixels) provide thorough description of flowfield.
- Eliminates ambiguities associated with interpreting 2-D data sets.
- Allows determination of directional information of gradients and correlations.
- Gives quantitative data on asymmetries of flowfield structures.

Figure 9. Three-dimensional fluorescence imaging and cube display of 3-d data.

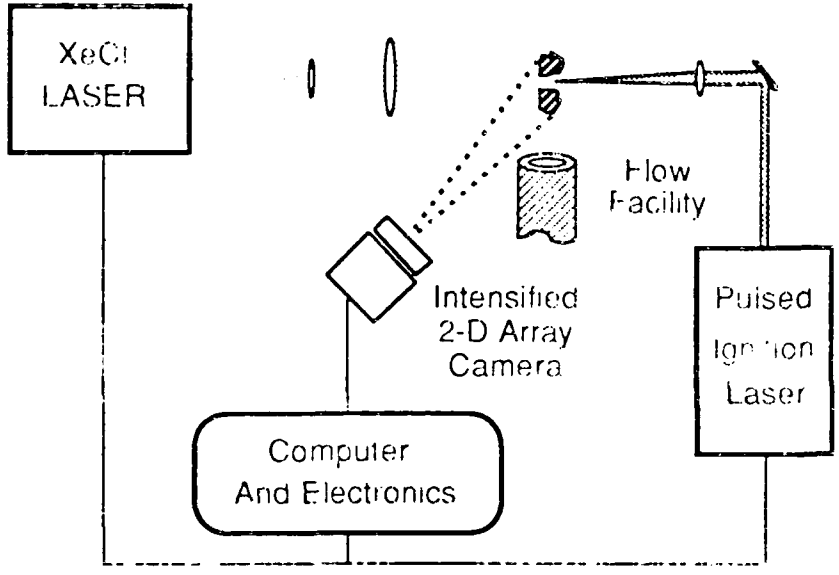
Another example of 3-d fluorescence imaging is provided in Fig. 10. In this case the quantity monitored is the OH concentration at a fixed time (0.5 milliseconds) after a pulse of focussed excimer laser radiation (193 nm) is used to ignite a flowing stream of premixed CH_4 and air at atmospheric pressure. The power of PLIF imaging in establishing a realistic model of the flowfield was readily apparent in this study, as the measurements revealed a much different OH distribution than was expected. Although we expected to observe a spherically expanding flame front, it is clear that the flame kernel propagates with cylindrical symmetry. In this case the OH probe beam was our recently acquired tunable excimer set to overlap a strong OH transition at 308 nm. This is the first example in our laboratory of pump-probe laser experiments in which an energetic laser pulse is used to intentionally perturb a combustible mixture while a second, weaker laser is used to probe the system via PLIF imaging. This general approach should have considerable value in future research.

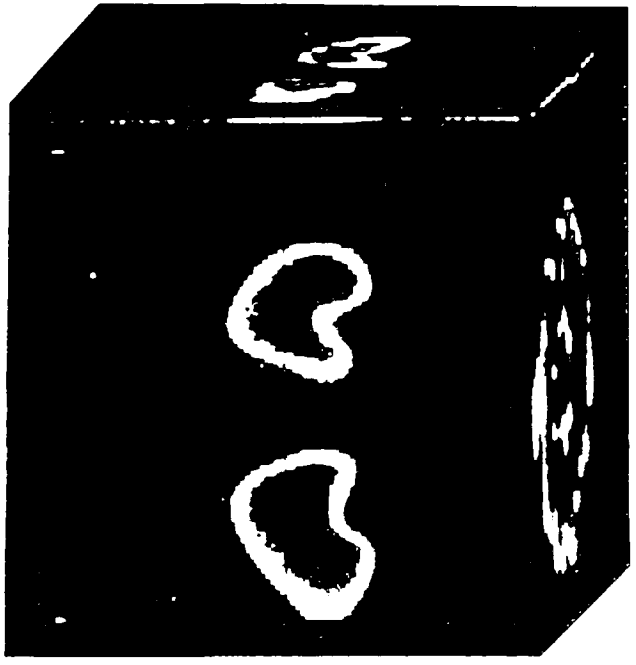
Our plans for continued research with 3-d imaging will involve: extending measurements to other flows and variables (e.g., O_2 concentration, temperature and particle size distributions), increasing the recording rate, and work with the Pixar computer to establish techniques for interactive display and processing of 3-d data.

(h) Velocity and Pressure Imaging

Velocity is a primary variable for many fluid mechanics studies, and it is typically measured by single-point methods. Clearly, an imaging diagnostic which could provide simultaneous multiple-point velocity data would have significant impact in many research areas involving fluid flow. Over the past few years we have worked to develop such a diagnostic method based on a variation of PLIF which is sensitive to velocity through the well-known Doppler effect. In brief, the method is a form of molecular velocimetry in that the quantity sensed is the mean velocity of the molecules at the measurement location. Through use of a laser source which is narrower than the absorption line being probed, the effective absorption coefficient of the gas depends on the difference between the fixed laser frequency and the velocity-dependent absorption line center. The subsequent fluorescence yield (fraction of absorbed photons converted to fluorescence photons) is independent of velocity but depends on the local temperature and pressure. By recording fluorescence images with different laser orientations, all within a time short compared to flow changes, ratios of signals can be used to cancel signal dependence on all parameters of the problem except the Doppler shift.

RAPID 3-D IMAGING ALLOWS TEMPORAL AND SPATIAL RESOLUTION OF COMBUSTION





3-D Display of OH Distribution
After Laser Ignition

- 120 000 Data Points in 3D Array Measurable in 0.05 seconds
- Captures Real-Time Ignition Phenomena

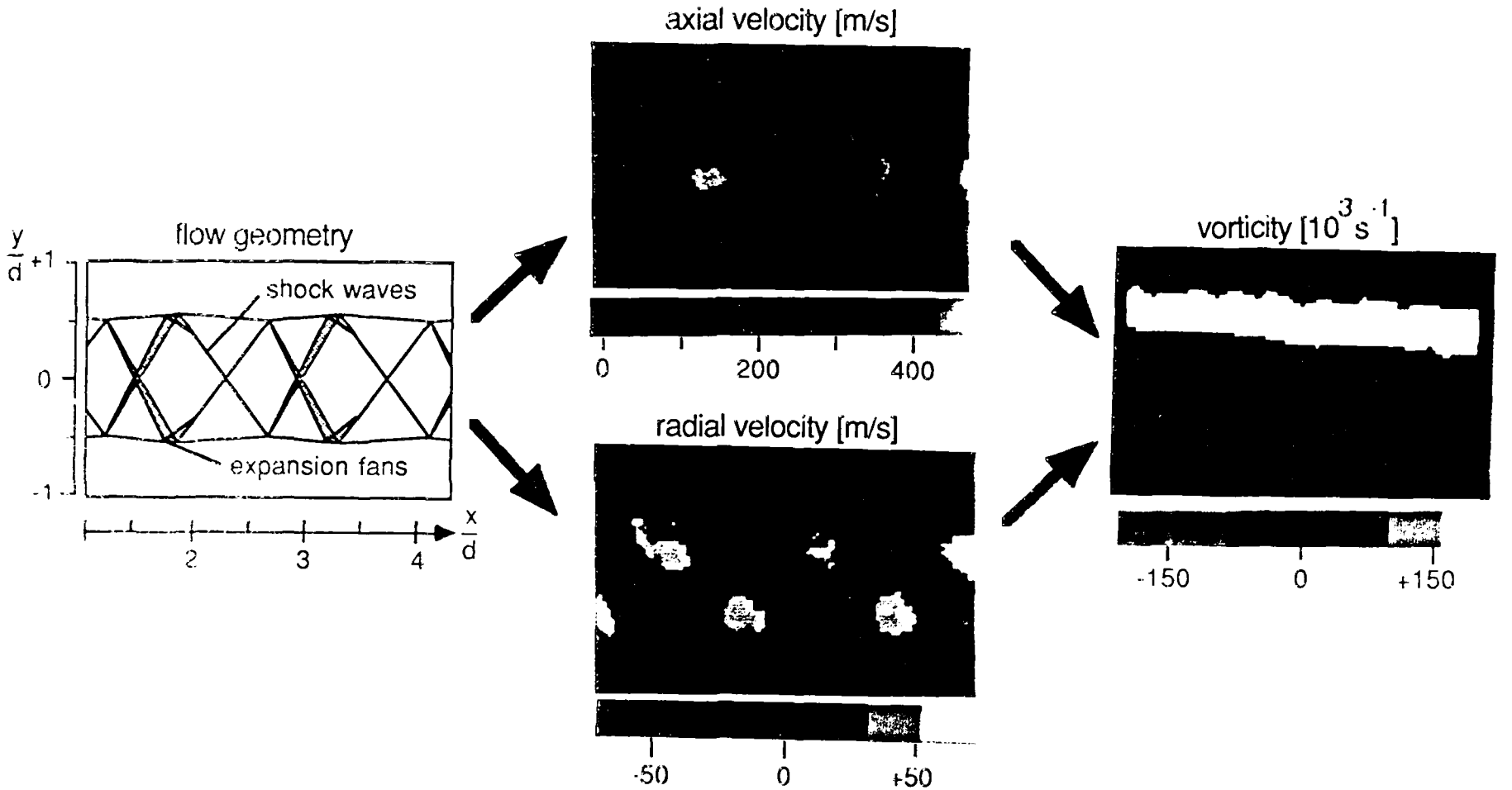
Figure 10. Application of 3-d fluorescence imaging of OH to investigate flame structure following pulsed laser ignition of flowing CH_4/air system.

Our work has been thoroughly documented (see papers 5, 6, 17, 34, 38 and 44) as we've progressed toward an optimum measurement strategy, and so no details are presented here. The most important points to note are that the laser source utilized was a tunable, single-mode cw argon ion laser (514.5 nm), which has a fortuitous coincidence with a pair of strong iodine transitions, and that the method has been demonstrated to work over a range of velocity from about 10 m/sec to several hundred meters per second. Indeed, the concept works best for high velocity flows, and this raises hope for a non-intrusive, multiple-point velocity diagnostic suitable for probing supersonic and hypersonic flows. We've now concluded our work with iodine-seeded flows and are turning our attention to high-speed air flows. An indication of our final capability with iodine-seeded nitrogen flows, for the case of an underexpanded supersonic jet, is shown in Fig. 11. The scheme employed (see papers 34 and 44) utilized four successive sheets of laser light, from a total of three different angles, allowing determination of two velocity components (in a central plane of the jet) and also the pressure from the measured local linewidth. Once the velocity field is known, of course, the vorticity can be computed, and a false-color of the vorticity field so inferred is shown in the figure. This is apparently the first vorticity image obtained without the introduction of particles into the flow.

With regard to current work, we are working to extend the molecular velocimetry concept to supersonic and hypersonic air flows by probing O_2. Although the measurement concept is similar, there are significant technological challenges to overcome in providing a narrow-linewidth source in the proper frequency region (i.e., the UV or VUV). The laser must be pulsed to operate in this spectral range, and it must have a sufficiently narrow linewidth relative to the absorption linewidths and Doppler shifts involved. In addition, since the repetition rate of most pulsed lasers is low, we must find a way to make the required illumination sheets from a single laser pulse or from synchronized, closely spaced pulses from multiple lasers. Finally, it will be necessary to record the multiple images in a short time, thereby placing severe constraints on the camera system employed.

The status of our work is that we are carrying out computer analyses of candidate schemes in order to select the optimum approach to be pursued in laboratory work. In parallel with this effort, we are assembling a small-scale supersonic flow facility in which measurements will be made, and we are performing modifications to our tunable narrow-linewidth excimer laser which may yield the necessary combination of laser energy and linewidth. We are also pursuing an alternate laser strategy, based on amplifying the Raman-shifted output of a tunable, narrow linewidth dye laser, to obtain even narrower

SUPERSONIC FLOW VELOCITY AND VORTICITY IMAGED BY LASER-INDUCED FLUORESCENCE



- First 2-D Vorticity Images in Gaseous Flows
- Particle Seeding Not Needed - Doppler-Shift at Molecular Level
- Application to Hypersonic Vehicles

Figure 11. Planar fluorescence imaging of velocity, pressure and vorticity in supersonic flows.

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linewidths. According to our calculations the latter scheme will provide a nearly optimum laser source for O_2 velocimetry; the shortcoming of this approach being the complexity and cost of the laser system. As part of this same analysis, we are pursuing promising schemes for simultaneous determination of O_2 concentration, temperature and pressure in compressible, reacting flows. Although the challenges are severe, the potential impact of a successful imaging diagnostic for nonreacting and reacting air flows clearly justifies a strong effort on this topic.

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Presentations

1. G. Kychakoff, K. Knapp, R. D. Howe and R. K. Hanson, "Quantitative Flow Visualization in Combustion Gases", paper No. 82-60, presented at Fall 1982 meeting, Western States Section/The Combustion Institute, Livermore, CA. (October 1982).

2. J. C. McDaniel, "Velocity Measurements Using Doppler-Shifted Laser-Induced Iodine Fluorescence," presented at Thirty-Fifth Meeting of the American Physical Society, Div. of Fluid Dynamics, Rutgers U., Nov. (1982).

3. G. Kychakoff, R. D. Howe, R. K. Hanson and K. Knapp, "Flow Visualization in Combustion Gases," paper 83-0405, presented at AIAA 21st Aerospace Sciences Meeting, Reno (January 1983).

4. J. C. McDaniel, "Quantitative Measurement of Density and Velocity in Compressible Flows Using Laser-Induced Iodine Fluorescence," paper 83-0049 presented at AIAA 21st Aerospace Sciences Meeting, Reno (January 1983).

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6. B. Hiller, J. C. McDaniel, E. C. Rea, Jr., and R. K. Hanson, "Laser-Induced Fluorescence Technique for Velocity Measurements in Subsonic Flows," presented at CLEO '83, Baltimore (May 1983).

7. G. Kychakoff, R. D. Howe and R. K. Hanson, "Use of Planar Laser-Induced Fluorescence for Combustion Measurements," presented at CLEO '83, Baltimore (May 1983)

8. G. Kychakoff, R. D. Howe and R. K. Hanson, "Quantitative Flow Visualization Technique for Measurements in Combustion Gases," presented at the Ninth International Colloquium on the Dynamics of Explosions and Reactive Flows, Poitier, France (July 1983).

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23. R. K. Hanson, J. M. Seitzman, J. Haumann, M. P. Lee and P. H. Paul, "Laser Fluorescence Imaging Diagnostics for Gaseous Flows," paper WG3 at Lasers '85, Las Vegas, Dec. 2-6, 1985.

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25. R. K. Hanson, "Laser Fluorescence Imaging in Spray Flames," invited speaker, General Motors Research Lab Colloquium, Warren, MI., Dec. 13, 1985.

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33. G. Kychakoff, P. H. Paul, I. van Cruyningen and R. K. Hanson, "High-Resolution and Three-Dimensional Digital Imaging of Jets," paper WSS/CI 86-15 at fall meeting of Western States Section, The Combustion Institute, Tucson, Oct. 27-28, 1986.

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7. J. C. McDaniel and R. K. Hanson, "Quantitative Visualization of Flowfields Using Laser-Induced Fluorescence," in *Proceedings of 3rd International Flow Visualization Symposium*, Univ. of Michigan, pp. 113-117, 1983.

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2.2 Laser Wavelength Modulation Spectroscopy

Introduction

Continuing improvements in tuning rates of narrow-linewidth laser sources offer opportunities for establishing new diagnostic techniques based on wavelength modulation concepts. Laser wavelength modulation spectroscopy refers to laser absorption or laser-induced fluorescence measurements carried out with a rapid-tuning single mode laser. This method involves quickly scanning a tunable cw laser across one or more isolated absorption transitions and recording the spectrally resolved absorption line profile using either absorption or fluorescence detection. The method is generally applicable to both infrared and UV/visible transitions, and it is particularly attractive for measurements in combustion gases and plasmas.

A primary advantage of wavelength modulation is that it provides a simple means of discriminating against continuum extinction and luminosity effects which can seriously hinder conventional laser absorption or fluorescence measurements in two-phase combustion flows and high-luminosity plasmas. Moreover, recording the fully resolved absorption line eliminates the need for uncertain linewidth assumptions in converting measured absorption (or fluorescence) to species concentration or temperature. Over the past several years, in AFOSR-sponsored work, we have demonstrated the utility of the wavelength modulation concept for combustion measurements, first using a commercially available rapid-tuning infrared diode laser to probe IR-active species, and more recently using a Stanford-built rapid-tuning dye laser to probe UV-active species.

Unfortunately, rapid-tuning dye lasers, needed for accessing a variety of important radical species which absorb in the near UV and visible, are not commercially available. Recognizing the importance of such a capability, we developed (under AFOSR support, see paper 1 in list at end of section) a novel and simple modification to a commercial ring dye laser which increases the scan repetition rate by three orders of magnitude (from about 4 Hz to 4 kHz) for short scans (up to 5 cm^{-1} = 150 GHz), and more recently we incorporated intracavity frequency doubling into the dye laser to permit access to UV wavelengths (see paper 6). Operation in the UV is critical for access to a variety of important combustion and plasma species.

Scientific Merit

Our laboratory now has unique capability and experience with fast-tuning lasers (UV, visible and IR) and wavelength modulation spectroscopy. This capability has provided several opportunities for pioneering contributions to combustion and plasma diagnostics research. In connection with combustors where particulates or droplets are present, wavelength-modulation techniques can be applied to discriminate between the gaseous absorption or fluorescence of interest and interfering continuum extinction. For plasma flows which may be highly luminous, wavelength modulation should provide a means of distinguishing the spectrally varying signal of interest from the intense continuum background. In unsteady flows or in devices where transient phenomena are of interest, fast measurements of fully-resolved absorption or fluorescence lines can be used for time-resolved determinations of species and temperature. Finally, the significance of fast-scanning capability for fundamental spectroscopic measurements should be noted. For example, during this program we have demonstrated the first measurements of fully resolved absorption lines in shock tube flows. Such experiments provide unique capability for obtaining a variety of fundamental high-temperature data including important quantities such as collision linewidths, oscillator strengths and heats of formation; these parameters are needed to enable quantitative absorption and fluorescence measurements of species in combustion and plasma flows.

Status Report

During this reporting period we have completed the laboratory portion of a multi-year study of wavelength modulation spectroscopy in OH. This work made use of the fast-tuning, frequency-doubled cw dye laser described previously (see paper 6) to probe various rovibronic transitions of OH in the (0,0) band near 300 nm. Two types of measurements were carried out: high-speed temperature measurements, and high-resolution lineshape measurements. The work was carried out using both a shock tube (see papers 8-11) and various laboratory flames (papers 10 and 12). Completion of the remaining data analysis and final publication of the results are expected within the next six months. The primary accomplishments of this project, which constitutes the Ph.D. theses of Ed Rea and Albert Chang, will be a new, fast temperature diagnostic for flames and shock tube flows, and significant contributions to the fundamental spectroscopic data base for OH.

Current work involves the extension of laser wavelength modulation spectroscopy to a new spectral region in the UV, namely 330-338 nm, which encompasses the strongest high temperature transitions of NH. This radical is of interest in the chemistry of combustion of nitrogen-containing fuels, but little or no work has been done to determine the collision-broadening parameters needed in quantitative determinations of NH by laser absorption and fluorescence methods. Our initial effort was concerned with modifying our cw dye laser to provide output in this spectral region, since commercially available ring lasers do not typically function in this range. We've tried and successfully demonstrated both intracavity and external frequency doubling, and have selected the latter approach as simpler to operate. A schematic of the optical arrangement using a lithium iodate crystal (angle-tuned phase matching) is shown in Fig. 12. The typical single-mode output power level achieved is a few microwatts, which is sufficient for the flame and shock tube measurements of interest.

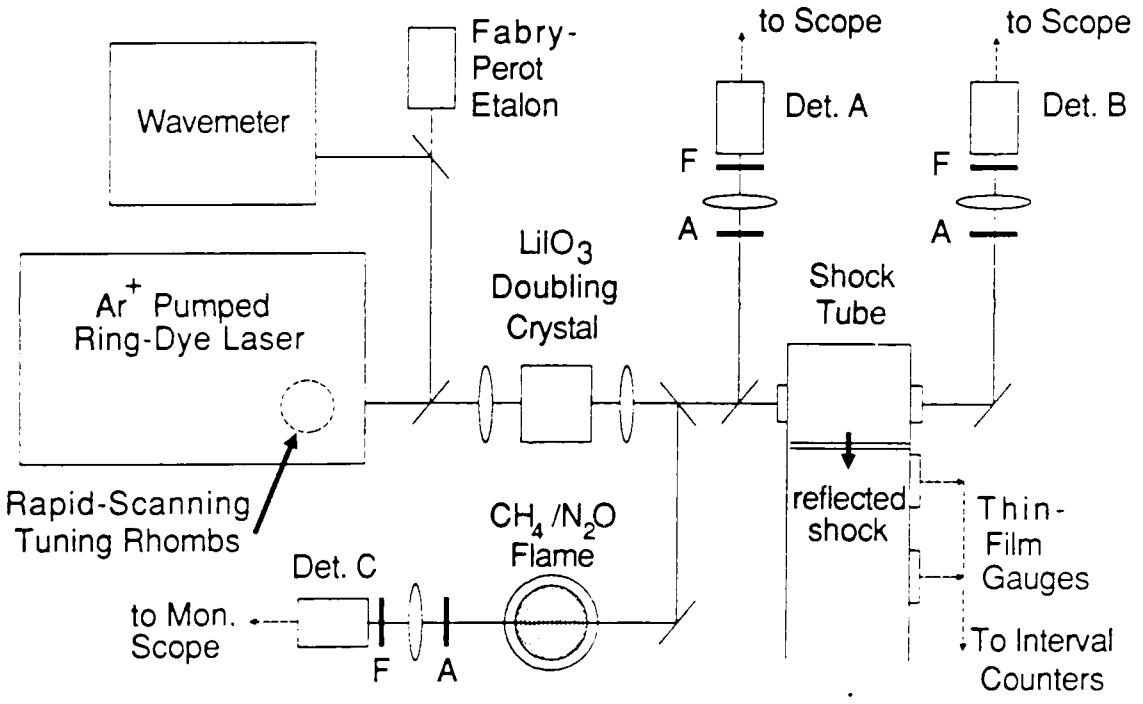

Figure 12. Schematic diagram for fast-scanning dye laser absorption experiments using external frequency doubling.

Once the laser was functional, we proceeded to carry out spectral surveys of NH absorption to validate our computer-based spectroscopic model. A sample result is shown in Fig. 13, which is a composite of 5 shock tube experiments in each of which the laser was scanned over a range of about 0.02 nm. The experiments were carried out at 2500 K in dilute NH_3-Ar mixtures, and the scans were made over a time interval when

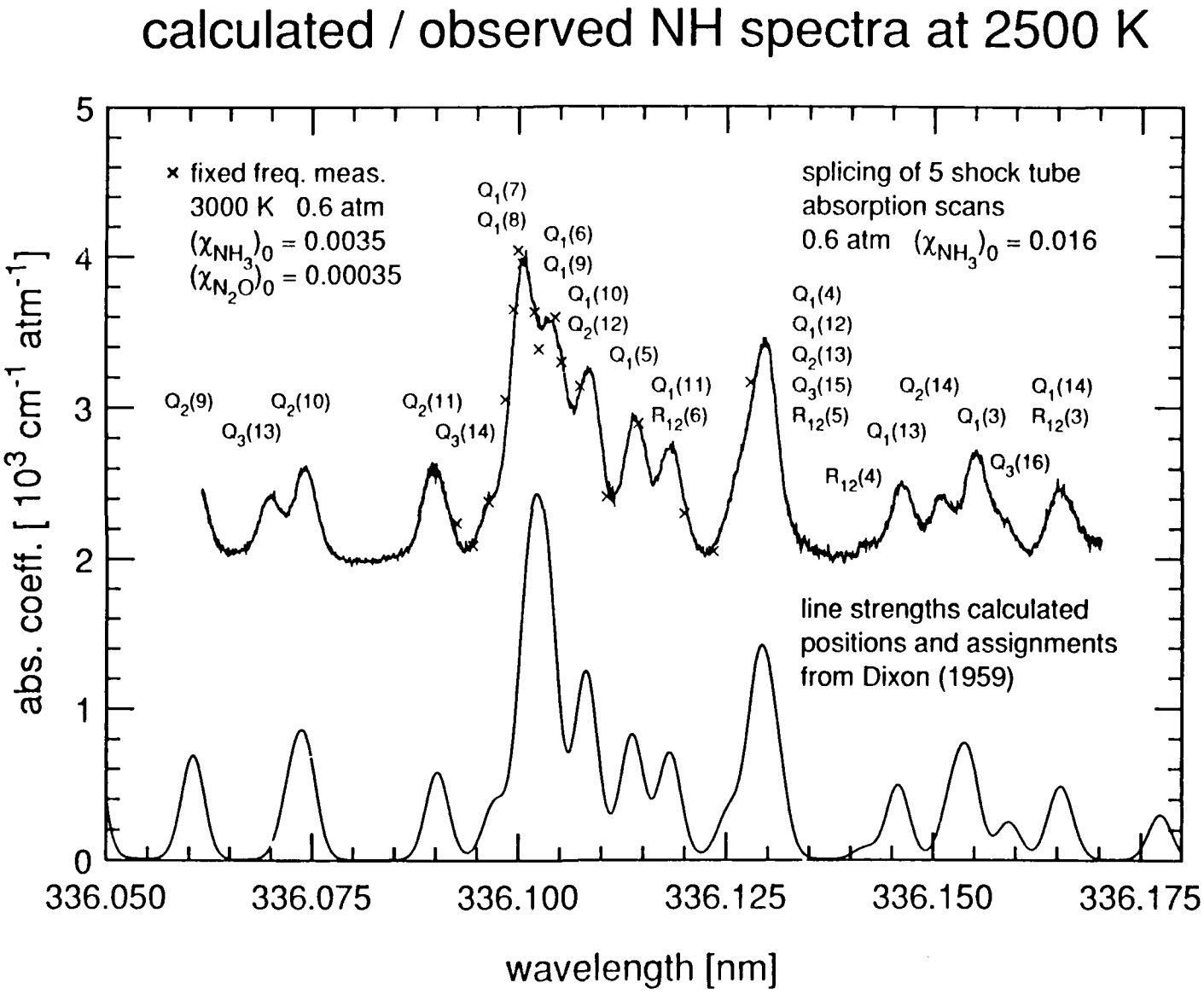

Figure 13. Spectral survey of NH absorption at 2500K obtained using laser-wavelength modulation spectroscopy in shock-treated NH_3-Ar mixtures.

the NH concentration was relatively constant. Inspection of the computed and measured spectral profiles reveals that the computer model accounts for the major features of NH, but that some minor features are missing and must be added to the model for completeness. An example of our initial work to determine line-broadening parameters is shown in Fig. 14 which is a high-resolution scan of the $Q_2(9)$ line near 336.05 nm. The best-fit value of the collision width (FWHM) per atmosphere of Ar pressure is 0.020 cm^{-1}-atm^{-1} at 2440 K. This may be compared with a value of about 0.025 for OH with a similar rotational quantum number. We expect to continue and complete our work on linewidths in NH during this next year.

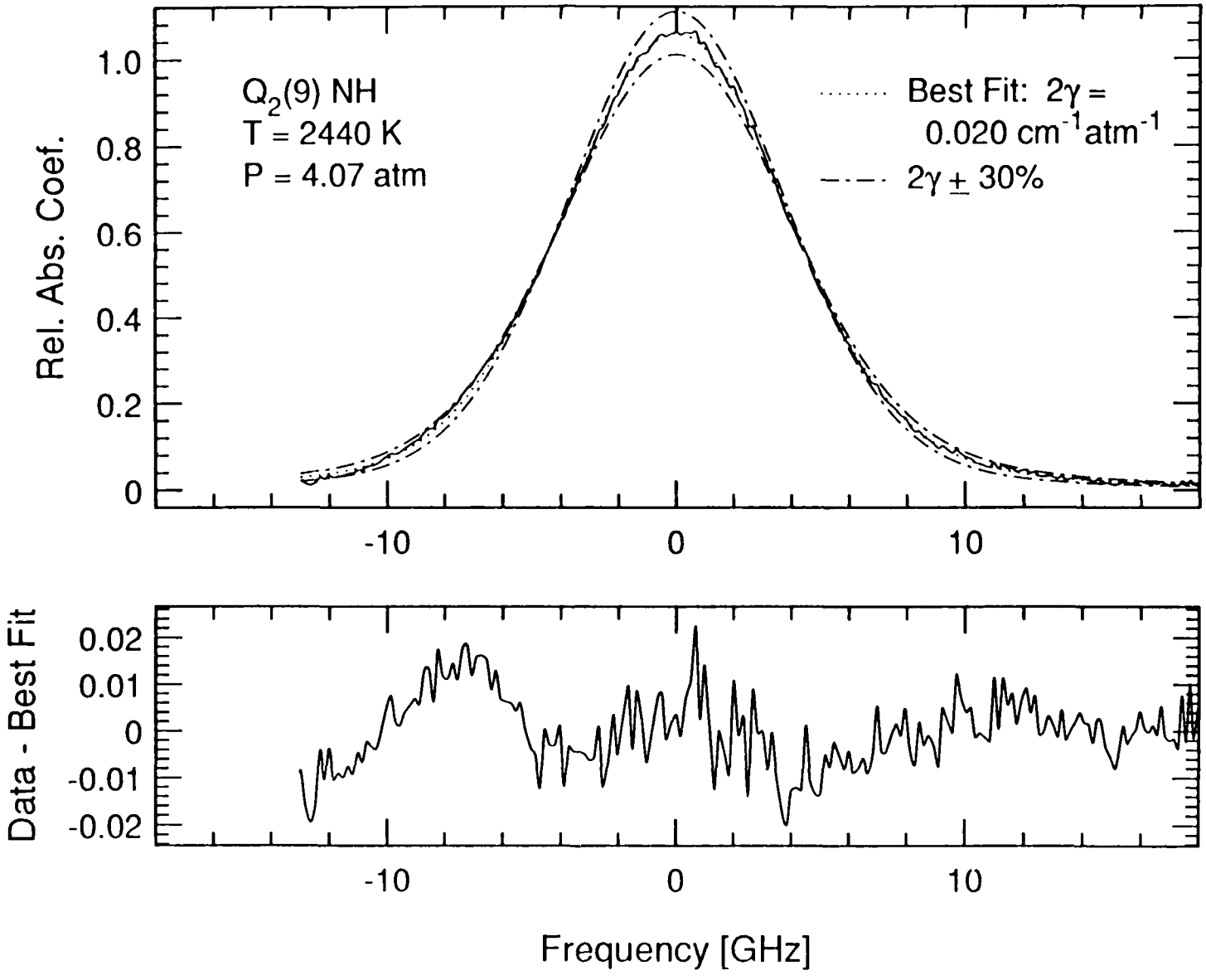

Figure 14. Fully resolved absorption lineshape in NH with best Voigt profile fit. Data obtained in shock-heated NH_3-Ar at 2440K.

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2.3 Plasma Diagnostics

Introduction

A relatively new element of our program is research on diagnostic techniques for plasmas. Primary motivation for this work arises from renewed interest in advanced space power and propulsion systems which may involve plasmas. Among the systems under consideration are thermionic converters and MHD generators for electrical power generation, MPD thrusters and beamed laser energy for propulsion, and direct production (in space) of high-power laser radiation for beamed energy. Considerable research will be needed before optimum systems for space utilization are identified, developed and placed in service, and we believe that advanced diagnostics will play an important role in such research. Furthermore, work on plasma diagnostics forms a logical and efficient extension of our current program.

Our effort during the past two years has been divided between diagnostics research for low pressure plasmas and atmospheric pressure plasmas. With regard to low pressure plasmas we have assembled a RF-powered, parallel plate discharge facility and have carried out a number of emission and fluorescence experiments in silane (SiH_4) plasmas. Our effort on atmospheric plasmas has been split between facility development and analysis of candidate diagnostic schemes, as reported below.

Scientific Merit

This research seeks to provide new diagnostic methods for use in studies of plasma properties and plasma phenomena. The two primary techniques which we are pursuing, namely PLIF imaging and wavelength modulation absorption/fluorescence, are novel and will provide unique capabilities for measurements in ionized gases. We intend to coordinate this work closely with other OSR-sponsored work on plasma sciences underway in our laboratory, and we expect the scientific merit and relevance of the work to be enhanced by these interactions.

Status Report

During this reporting period we have completed two studies of low pressure silane plasmas and have participated in the installation of a major new atmospheric plasma facility for our laboratory. The low pressure work involved design and assembly of a RF-powered plasma reactor and subsequently the development and use of spectroscopic diagnostics to probe the spatial distribution of SiH in the chamber. We chose silane as a working material because of its relevance to semiconductor processing, and then focussed on SiH for diagnostics studies owing to its potential in process monitoring schemes and its reasonable spectroscopic simplicity. The diagnostics work involved both electronic emission, which allows measurements of excited state populations (SiH*) and laser-induced fluorescence, which monitors the ground state SiH concentration. In both cases the spectral region is near 413 nm, and the measurements made provided spatial distributions of concentrations across the gas between the parallel plate electrodes. The discharge conditions were typically 0.1 Torr of undiluted silane or diluted silane-argon mixtures; the electrode spacing was typically 25 mm and the electrode temperatures varied up to 320°C. Details of the work are given in papers 1 and 2 listed at the end of this subsection.

Our major activity during the past year has been to participate in the installation and initial testing of a major new plasma torch facility for our laboratory. The 50 kW,

inductively heated torch was purchased with an AFOSR DOD equipment grant (Prof. C. H. Kruger, Principal Investigator). Professor Kruger's primary interest is in basic studies of plasma nonequilibrium phenomena, but he has agreed to make the torch available for our diagnostics research. The tools which we develop, of course, will also be available to him and his students in their research.

The status of the torch is that is now installed and undergoing tests to map out the operating range for various feed gases. Most testing has been done with argon, though a few tests with hydrogen added at low levels have confirmed that a discharge can be sustained in a hydrogen-bearing mixture. This is significant since one of our primary diagnostics objectives is to monitor electron number density through measurements of Stark-broadened linewidths of atomic hydrogen. Spectral scans of plasma emission have been carried out and these enable determination of the plasma temperature at the torch exit. Analysis of these data using an Abel inversion to account for the radial distribution of the plasma properties leads to peak temperatures of about 9000 K.

With regard to diagnostics research, we have identified several goals which appear feasible based on calculations. Initially we plan to measure H-atom concentration profiles is the torch using two-photon-excitation absorption and fluorescence. The relevant energy levels of H are shown in Fig. 15, and a schematic of the proposed experimental arrangement for PLIF imaging is given in Fig. 16. Our plan is to use two photons at 205 nm, provided by Raman shifting (in H_2) an ArF laser (at 193 nm), to excite the $n = 1 \rightarrow 3$ transition of H. In the case of fluorescence we will monitor the subsequent emission at 656 nm to avoid interference with scattered light at the laser wavelength. Both our broadband and tunable narrowband excimer lasers are candidate sources. The optimum choice depends on how we plan to deal with the Stark-broadened lineshape of H. For H concentration measurements alone, it may be simpler to use a broadband laser; the tunable laser, however, may enable combined concentration and electron density measurements.

The status of the diagnostics research is that the Raman shifter is currently being assembled to generate the 205 nm light needed for H measurements. We are also carrying out an analysis of similar schemes for O-atom measurements, for which two-photon excitation at 226 nm appears attractive. Finally, we are considering measurements of NO (one-photon excitation at 226 nm), O_2 in the range 193 to 248 nm, and electron density. Most of these measurements require the same equipment and will be pursued in the laboratory once the torch is fully characterized and the Raman shifter is operational.

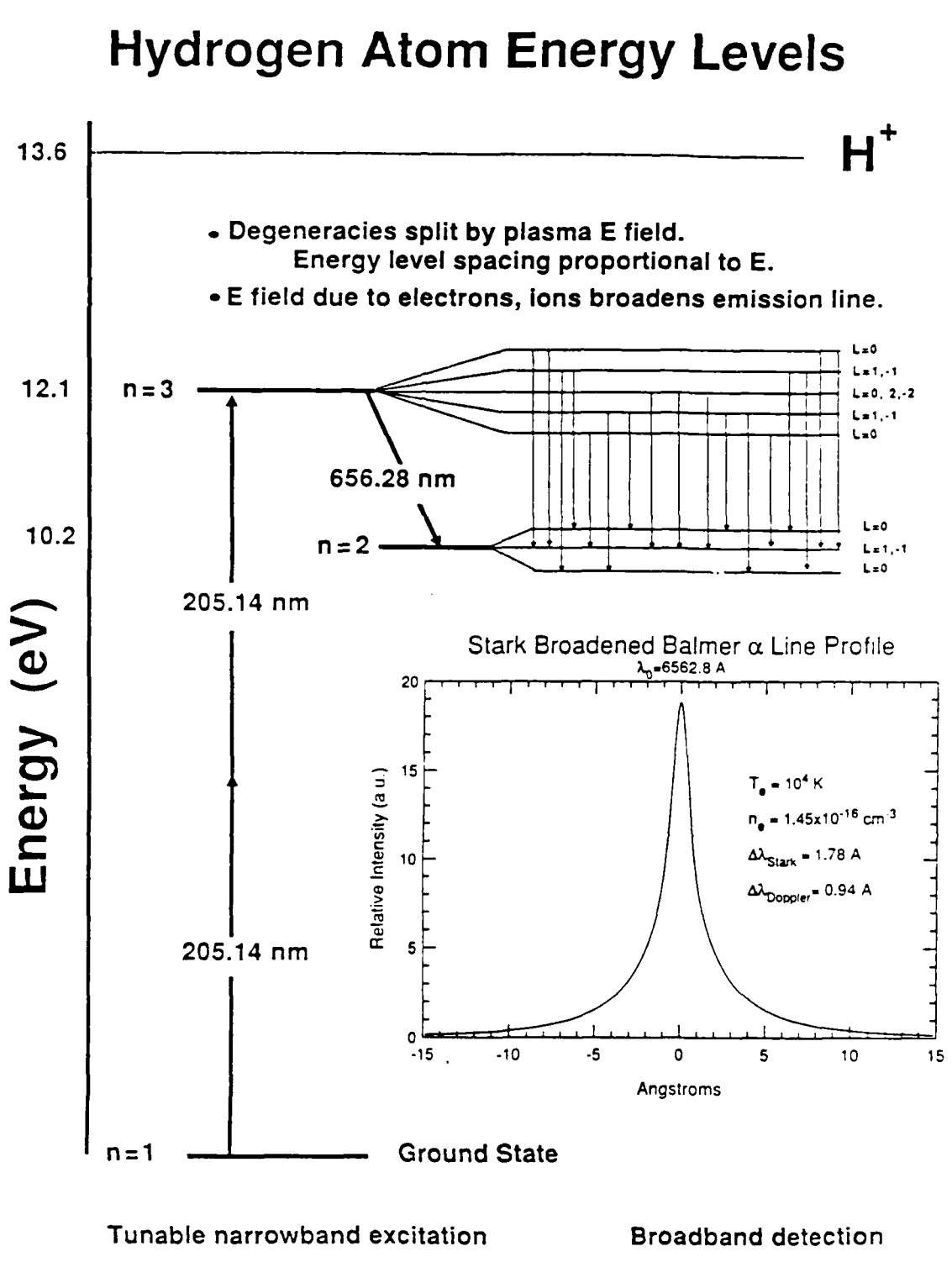

Figure 15. Energy level diagram for H indicating two-photon excitation path.

Experimental Setup

2 Photon Digital Imaging of H Atoms

- Tunable narrowband excitation
- Broadband detection

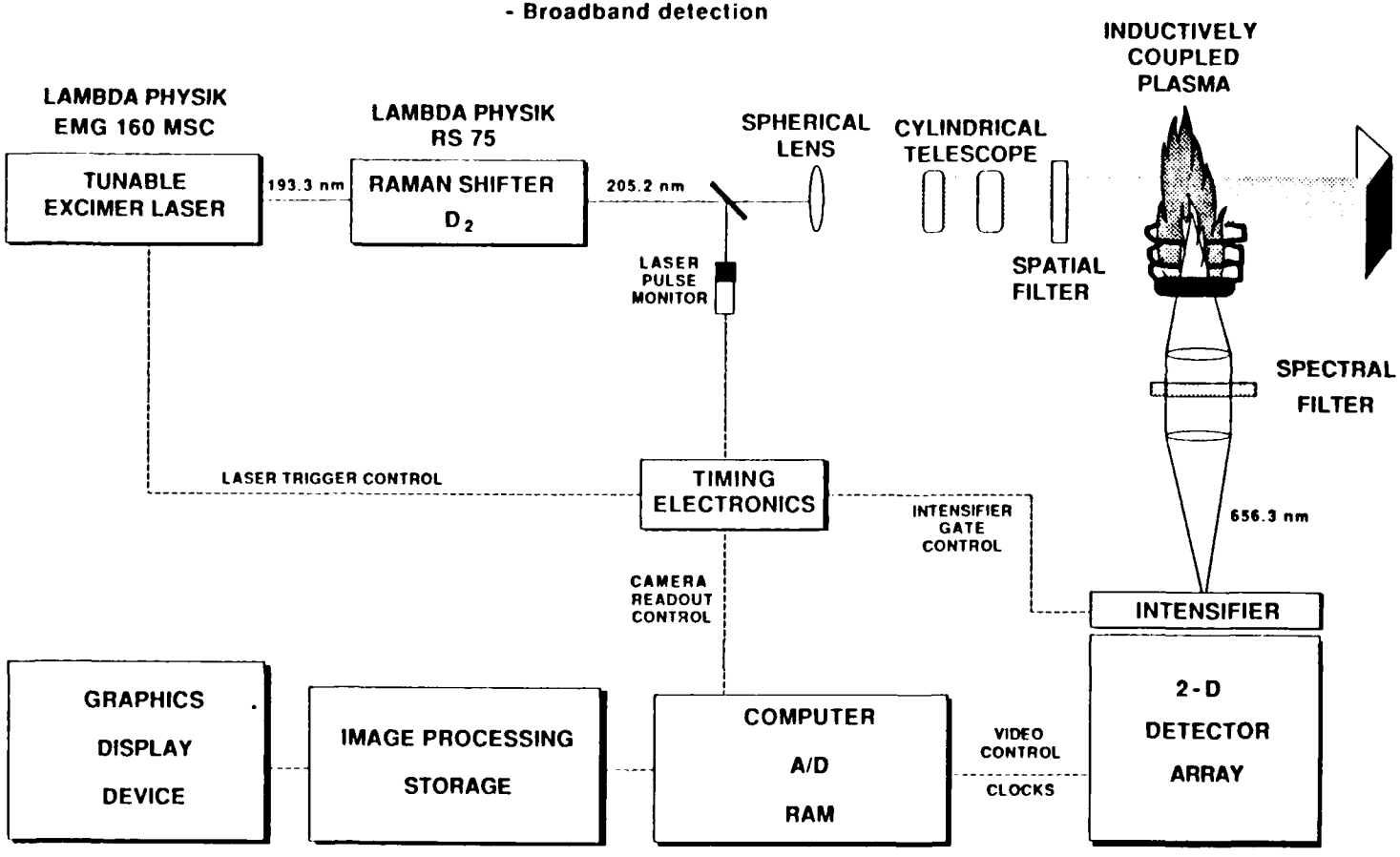

Figure 16. Schematic diagram of proposed two-photon PLIF imaging of H in a plasma torch.

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2.4 Laser Photolysis Shock Tube

Introduction

Our diagnostics work with excimer lasers has made us aware of the possible influence of high energy UV illumination on the chemical composition of gaseous systems. Although changes in composition represent an interference effect in some diagnostics applications, it should be possible to use these same effects to advantage in fundamental studies of the kinetics and spectroscopy of radical species. In fact, chemists have recognized this for several years, and there are an increasing number of studies being reported on the use of excimer laser photolysis to generate controlled samples of radical species. Thus far the application of laser photolysis has been limited to relatively low temperatures (in fact most often the experiments are conducted at room temperature), but it occurred to us a few years ago that it should be possible to combine the attributes of shock tubes (e.g., the ability to generate controlled temperatures to at least 5000 K) and

laser photolysis to enable more direct studies of radical species at high temperatures than was previously possible. We are now prepared to report our initial work in this direction.

Scientific Merit

The proposed concept, namely the direct and immediate production of controlled levels of radical species in high temperature gases by photolytic means, offers prospects of the first direct studies of various radical-radical and radical-molecule reactions at high temperatures. There are a number of such reactions of significance to the scientific community at large and to the Air Force in particular, and the concept in question should enable more accurate determinations of the reaction rate coefficients than is possible by other means. A related scientific opportunity is to study fundamental spectroscopic parameters of these radicals at elevated temperatures; the previous difficulty in generating controlled levels of these species has been a major handicap to spectroscopists interested in such properties. Finally, we note that the quantum level specificity of both excitation and probe lasers will eventually lead to controlled studies, in the high temperature environment provided by the shock tube, of detailed state-to-state chemistry and energy transfer which have not been previously studied.

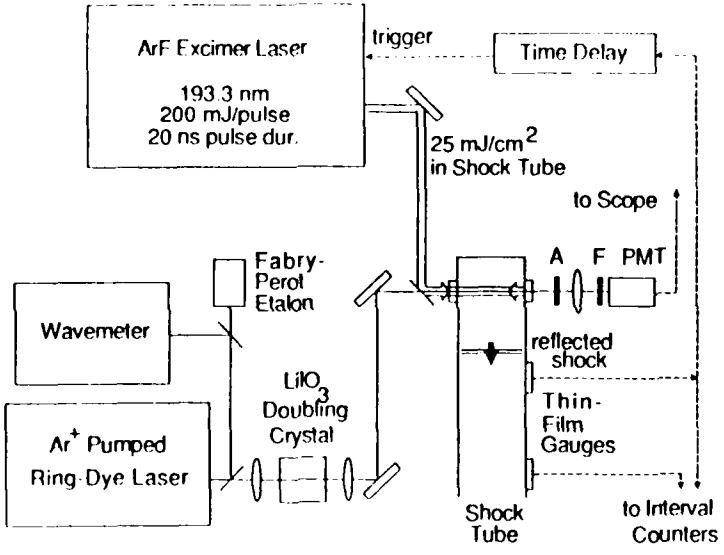
Status Report

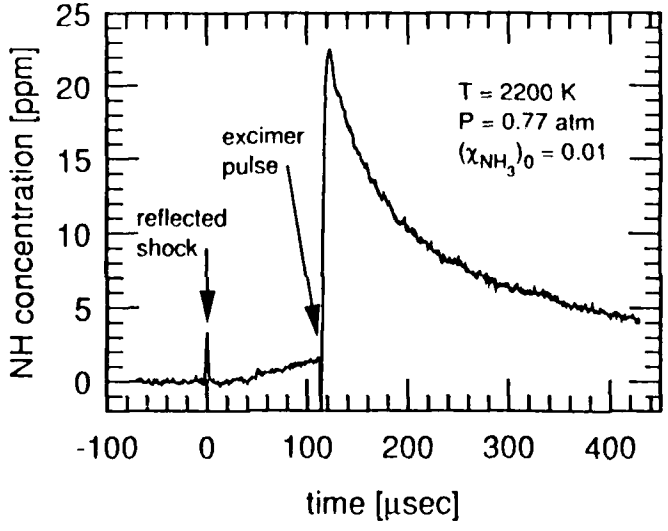
During the past year we have set up a new pressure-driven shock tube which will enable both conventional shock tube and laser-photolysis shock tube studies. Primary support for the facility has been provided by AFOSR, though some elements of the system have been funded by DOE. The facility is now operational though some problems with electrical interference (from the laser power supply) and with the shock velocity detection system still remain to be solved.

A schematic diagram of the facility as now configured is shown in Fig. 17. A Questek 2020 laser is used as the photolysis source, and thus far we have limited our work to ArF (193 nm) as the lasing medium. The optical arrangement under study is transverse illumination with a co-propagating cw laser probe beam. We have selected NH_3 as the first precursor species owing to our interest in the kinetics of NH_i compounds and the fact that NH_3 is known to have a large photodissociation cross-section at 193 nm. At present we are conducting tests to select optical components which hold up under the intense UV illumination provided by the focussed excimer output, and we are beginning measurements of the absorption coefficient of NH_3 at 193 nm as a function of temperature; this latter information is critical to planning experiments and is not available in the

LASER PHOTOLYSIS SHOCK TUBE FOR FUNDAMENTAL STUDIES OF COMBUSTION RADICALS

- EXCIMER PHOTOLYSIS PROVIDES CONTROLLED POOL OF RADICALS IN SHOCK-HEATED GASES





- PROVIDES FIRST-TIME ACCESS TO MANY CRITICAL COMBUSTION SPECIES
- ENABLES FUNDAMENTAL SPECTROSCOPIC STUDIES OF RADICALS AT HIGH TEMPERATURES
- ENABLES FUNDAMENTAL KINETICS STUDIES OF RADICALS AT HIGH TEMPERATURES

Figure 17. Schematic diagram and sample results for laser photolysis production of NH in a shock-heated NH_3-Ar mixture at 2200K.

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literature. Some preliminary experiments demonstrating that the ArF does indeed create radicals in NH_3 have been done, however, to build confidence in our approach. An example result is shown in the right-hand panel of Fig. 17. The instantaneous incremental production of NH is clear. Of course the primary photolysis products are NH_2 and H, but we do not yet have a detection scheme set up to monitor NH_2. Initial attempts to compute the NH time history using a standard NH_3 decomposition reaction mechanism have shown good agreement with the data.

In summary, we have confidence that this new approach to high temperature studies of radical species will provide an important new experimental tool for fundamental studies of the properties of high temperature gases. This same experimental facility should provide an excellent test bed for the development of advanced optical diagnostic techniques for high temperature gases and plasmas.

Presentations and Publications

None

Personnel

Ronald K. Hanson	Professor, Mechanical Engineering
David Davidson	Research Associate, Mechanical Engineering
Albert Chang	Graduate Student, Mechanical Engineering (Ph.D. expected in December 1988)

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