

AD-A238 028

MENTATION PAGE

Form Approved
GSA No. 0704-0188

2



is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302, and to the Office of Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503.

1. AGENCY USE ONLY (Leave blank)		2. REPORT DATE May 9, 1991	3. REPORT TYPE AND DATES COVERED Final, January 1, 1988 - January 31, 1991	
4. TITLE AND SUBTITLE "Nonlinear Spectroscopy of Multicomponent Droplets and Two- and Three- Dimensional Measurements in Flames" (u)			5. FUNDING NUMBERS PE - 61102F PR - 2308 TA - A3 G-88-0100 AFOSR 88-0100	
6. AUTHOR(S) Richard K. Chang and Marshall B. Long			7. PERFORMING ORGANIZATION REPORT NUMBER AFOSR-TR- 91 0599	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Departments of Applied Physics and Mechanical Engineering YALE UNIVERSITY P.O. Box 2157 Yale Station New Haven, CT 06520-2157			8. PERFORMING ORGANIZATION REPORT NUMBER	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) AFOSR/NA Building 410 Bolling Air Force Base, DC 20332-6448			10. SPONSORING/MONITORING AGENCY REPORT NUMBER AFOSR-TR- 91 0599	
11. SUPPLEMENTARY NOTES				
12A. DISTRIBUTION/AVAILABILITY STATEMENT Approved for public release; distribution is unlimited			12B. DISTRIBUTION CODE	
13. ABSTRACT (Maximum 200 words) Significant progress has been made in the following two research areas: (1) Nonlinear spectroscopy of micrometer-size droplets; and (2) Development and application of two- and three-dimensional scalar measurement techniques in flames. In the nonlinear spectroscopy area, the following achievements are reported: (1) Laser-induced shape deformation of transparent droplets by electrostriction, which pushes against the surface tension force and causes the droplet to bulge; (2) Laser-induced shape deformation by heating of absorbing droplets, which were imaged with fluorescence photography; (3) Laser-induced breakdown which quenches the stimulated Raman scattering (SRS); (4) Excitation of SRS with single-mode and multimode Q-switched lasers; and (5) Four-wave mixing processes in droplets, such as third-order sum frequency generation. Achievements in multidimensional scalar imaging include the following: (1) Measurement of the time evolution in premixed H ₂ -air flames using imaging techniques; (2) Development of new techniques for following the time evolution of flow structures in three dimensions; (3) Introduction of a technique for visualizing supersonic flows using Rayleigh scattering from condensed-phase droplets; (4) Measurement of the complete scalar gradient in a nonreacting flow, which allows calculation of the scalar dissipation; (5) Simultaneous CH and CH ₄ mapping in a turbulent nonpremixed flame, and (6) Investigation of differential diffusion effects.				
14. SUBJECT TERMS droplets, stimulated Raman scattering, laser induced breakdown, laser-induced deformation, four-wave mixing, premixed flames, nonpremixed flames, Rayleigh scattering, three-dimensional measurements, laser diagnostics, flow imaging			15. NUMBER OF PAGES 23	
17. SECURITY CLASSIFICATION OF REPORT Unclassified			16. PRICE CODE	
18. SECURITY CLASSIFICATION OF THIS PAGE Unclassified		19. SECURITY CLASSIFICATION OF ABSTRACT Unclassified		20. LIMITATION OF ABSTRACT SAR

FINAL REPORT
to the
Air Force Office of Scientific Research

Nonlinear Spectroscopy of Multicomponent Droplets
and
Two- and Three-Dimensional Measurements in Flames

AFOSR Grant No. 88-0100

January 1, 1988 - January 31, 1991

Richard K. Chang and Marshall B. Long
(Co-Principal Investigators)

Yale University

New Haven, Connecticut 06520-2157

May 15, 1991

Assignment For	
DTIC Grant	<input checked="" type="checkbox"/>
DTIC Tab	<input type="checkbox"/>
Unannounced	<input type="checkbox"/>
Justification	
by	
Distribution/	
Availability Codes	
Disc	Avail and/or Special
A-1	

91-04739



Approved for public release; distribution unlimited.

91 7 11 033

CONTENTS

	<u>Page</u>
Introduction	1
Research Accomplishments	
I. Nonlinear Spectroscopy of Droplets	1
II. Multidimensional Imaging in Flames	7
Publications Resulting from the Research	15
Scientific Collaborators	17
Lectures Presented about the Research	18
Interactions with Other Laboratories	21

INTRODUCTION

During the three years of AFOSR support, significant progress has been made in the following two research areas: (1) Nonlinear spectroscopy of micrometer-size droplets; and (2) Two- and three-dimensional scalar mapping. Specific details of our accomplishments regarding nonlinear interactions in droplets and multidimensional diagnostics in flames can be found in the publications resulting from the research (see list starting on page 15). All these papers have been submitted to AFOSR in both preprint and reprint form.

RESEARCH ACCOMPLISHMENTS

I. Nonlinear Spectroscopy of Droplets

Following is a brief description of the three principal research objectives related to nonlinear spectroscopy of liquid droplets, as outlined in our original proposal:

1. To investigate the stimulated Raman scattering (SRS) statistics from single droplets in a flowing linear stream with the following three types of laser excitation: (1) a cw mode-locked Nd:YAG laser which can be pulsed at a rate of 80 MHz, thereby enabling us to average the SRS signal over many laser pulses per second of integration time; (2) a multimode Q-switched Nd:YAG laser which has a linewidth of 0.6 cm^{-1} and numerous picosecond spikes superimposed on the nominal 10 ns Q-switched pulse; and (3) a single-mode Q-switched Nd:YAG laser (with an injection seeder) which has a linewidth of 0.006 cm^{-1} and a temporally smooth pulse of 7-10 ns.
2. To explore the possibility of using CARS spectroscopy to determine the concentration distribution from different regions within the droplet rim. We plan to use a CCD two-dimensional detector to accumulate the CARS signals from numerous laser shots from a single-mode Q-switched laser or a cw mode-locked

laser. Particular emphasis will be placed on the nonuniform concentration distribution within a multicomponent fuel droplet which results from combustion, nonuniform heating, and acceleration of the droplet.

3. To determine to what extent the laser pulse deforms the transparent droplet via electrostrictive forces. Knowledge of such laser-induced shape deformation is important in determining the Q factor of the morphology-dependent resonances (MDR's) which provide the necessary optical feedback for SRS within the droplet.

A curved liquid-air interface of a micrometer-size droplet has the following unique characteristics: (1) enhances the internal intensity of the incident radiation; (2) enhances the spontaneous emission transition rates, such as the Raman and Brillouin scattering coefficients; and (3) provides feedback to the internally generated nonlinear waves. The enhanced internal intensity not only increases the nonlinear signals but also increases the probability of deforming the droplet shape and/or causing laser-induced breakdown (LIB). Consequently, the research of nonlinear spectroscopy of droplets necessitates the study of laser-induced shape deformation and LIB.

Following is a brief summary of the research accomplishments in several areas of nonlinear optical interactions with micrometer-size droplets:

(a) Laser-Induced Shape Deformation by Electrostriction

We have demonstrated that shape distortion of totally transparent droplets can result via the electrostrictive force associated with the gradient of the laser intensity (∇I) which is largest on the surface of the droplet shadow face. The laser-induced electrostrictive force pushes against the surface tension force of the droplet and causes the droplet to bulge at the shadow face. After several microseconds, the droplet shape oscillates between a spheroid and a sphere, the distortion amplitude finally dampens, and the droplet remains spherical in shape. The shape oscillation is proportional to the square root of the dynamic surface tension

of the liquid, and the damping rate is inversely proportional to the bulk viscosity of the liquid. Quantitative information on laser-induced shape distortion is important because this electrostrictive effect sets an upper limit on the amount of laser energy needed to shatter a droplet. Furthermore, knowledge of the electrostrictive effect enables us to estimate the shape distortion amplitude induced by a high intensity laser pulse, which is used to pump the SRS. [Our results are published in Optics Letters 13, 916 (1988), see publication #2.]

(b) Laser-Induced Shape Deformation by Heating

We have investigated the shape distortion of absorbing droplets after irradiation by a laser beam. We developed a fluorescence technique which can image the liquid (not the vapor) phase of a droplet and the ejected material, after a CO₂ laser pulse nonuniformly heats the droplet. The shapes of the parent droplet and of the ejected liquid material were studied after laser-induced explosive vaporization of water and ethanol droplets, which have large differences in their absorption at 10.6 μm and hence different material ejection characteristics. With this technique, we can visualize the parent droplet and the liquid phase portion of the ejected material without the interfering contribution from the vapor phase portion of the ejected material. After irradiation of the droplets with a CO₂ laser beam, the droplet shape distortion, ejection, shattering, and propulsion have been photographed using the fluorescence emission from rhodamine 6G dye added to water (1×10^{-4} M) and to ethanol (0.2×10^{-4} M). The fluorescence technique is a variation of an imaging approach initially developed by Prof. Lynn Melton to determine the internal temperature distribution within a droplet using exciplex-monomer fluorescence. [Our results are published in the Proceedings of a 1989 AGARD Conference and in Optics Letters 15, 664 (1990), see publications #5 and #6, respectively.]

(c) Laser-Induced Breakdown which Quenches SRS

At high input laser intensities, well above the SRS intensity threshold, the laser-induced breakdown of droplets sets another upper intensity limit which should not be exceeded. LIB is accompanied by the development of a dense, high temperature plasma

which can absorb the SRS. Such breakdown is localized in a region just within the droplet shadow face and is initiated during the rising portion of the laser pulse, which pumps the SRS. Optical absorption of the plasma quenches the SRS during the subsequent portion of the laser pulse. Consequently, the higher the input laser intensity is, the less SRS is generated once laser-induced breakdown occurs.

We studied the growth, decay, and quenching of SRS in transparent droplets using a time-resolved spectroscopic technique which consists of a spectrograph placed in front of a framing camera. The spectrograph disperses the radiation from the laser and the first- and j th-order Stokes SRS along the vertical axis, and the framing camera displays the temporal information along the horizontal axis. The time delay between the first-order Stokes SRS and the input laser pulse is the buildup time of the SRS starting from the spontaneous Raman noise within the droplet. The much shorter time delay between the first-order Stokes and the higher-order Stokes is indicative that, for the cascading stimulated Raman process, the initial Raman signal is produced by the four-wave mixing process, not by the spontaneous Raman process as is the case for the first-order Stokes SRS. The various orders of Stokes SRS radiation continue to circulate within the droplet, even after the laser pulse is off. The radiation decay rate is dependent on the leakage rate of the MDR's which provide the optical feedback for the SRS radiation and on the intensity-dependent depletion rate which results from the cascading SRS. The onset of LIB is sensitively indicated by the quenching of SRS, which precedes our ability to detect atomic emission from species within the laser-induced plasma. [Our results are published in *Optics Letters* **13**, 559 (1988) and the *Proceeding of Conference on Laser Materials and Laser Spectroscopy*, see publications #1 and #3, respectively.]

(d) Excitation of SRS with Single-Mode and Multimode Q-Switched Lasers

We completed the experimental study of the SRS statistics from single droplets in a flowing stream (our first research objective). We compiled our results in the form of a

histogram of the SRS intensity for 700 laser shots at a fixed intensity. The fluctuation of the SRS intensity for single-mode laser pumping was much less than that for multimode laser pumping. Therefore, single-mode laser pumping should always be used to minimize fluctuations in the SRS intensity.

The SRS intensity threshold was also investigated with single-mode and multimode Q-switched Nd:YAG laser excitation. The new findings can be summarized as follows: (1) The SRS intensity threshold with a single-mode laser beam is noted to be three times lower than that with a multimode beam. (2) The intensity threshold for stimulated Brillouin scattering (SBS) from droplets is lower than that for SRS with single-mode excitation. (3) both SBS and SRS appear as equal length arcs which are confined to the droplet illuminated and shadow faces; (4) the SRS and SBS consist of several pulses within the smooth Q-switched laser pulse (≈ 7 ns duration); (5) the first SBS pulse always occurs sooner than the first SRS pulse; and (6) the temporal profiles of the SRS and SBS pulses, which are simultaneously measured with a streak camera (100 ps resolution), are temporally correlated, i.e., the minimum of the $(n + 1)$ th SBS pulse occurs when the n th SRS pulse reaches a maximum.

Based on the SRS and SBS results, we conclude that the following processes occur within droplets upon irradiation by a single-mode laser beam: (1) the droplet illuminated face enhances the incident intensity in a region just within the droplet shadow face; (2) both spontaneous Brillouin scattering and Raman scattering are created and amplified with optical feedback provided by the droplet morphology; (3) the SBS threshold is exceeded before the SRS threshold; (4) the SBS, rather than the laser radiation, within the droplet serves as the pump for SRS; (5) the first SBS pulse is depleted by pumping the first SRS pulse; (6) the first SRS pulse is depleted by the cascade stimulated Raman process; (7) the second SBS pulse needs to be repumped by the remaining single-mode laser pulse; (8) the second SRS pulse needs to be repumped by the second SBS pulse; and (9) for the third SBS and SRS pulses, the previous sequences are repeated until there is not enough laser intensity to repump the n th SBS

pulse. The most striking finding from this phase of our research is that the SRS is pumped by the SBS, not by the laser pulse. [Our findings are published in the Journal of the Optical Society of America B 7, 108 (1990), see publication #4.]

(e) Four-Wave Mixing in Droplets

The second objective of our research is to explore the possibility of using CARS spectroscopy to determine the concentration distribution from different regions within the droplet rim. CARS is one example of the more general four-wave mixing processes which require phase-matching. Numerous phase-matching configurations (such as BOXCARS) have been conceived for plane waves in an extended medium. However, phase matching is poorly understood for waves circumnavigating the droplet rim. For example, what is the phase velocity of a MDR, which can be envisioned as two counterpropagating guided waves?

We initiated an experimental and theoretical program to further our understanding of the phase velocity of guided waves within droplets. We chose to use third-order sum frequency generation (TSFG) as a means of providing more direct information about the phase velocity of MDR's and, more specifically, about the phase-matching ability among waves trapped within the droplet. The nonlinear source polarization which induces the electric field at the third-order sum frequency is:

$$P^{NLS}(\omega_1 + \omega_2 + \omega_3) = \chi^{(3)} E(\omega_1) E(\omega_2) E(\omega_3),$$

where ω_1 , ω_2 , and ω_3 are the laser frequency at ω_L and/or the j th-order Stokes SRS frequency at ω_{js} . As in the case for the CARS intensity, the TSFG intensity is proportional to the square of the coherence length $(l_{coh})^2$, where $l_{coh} = \pi/\Delta k$. For the degenerate case (when $\omega_1 = \omega_2 = \omega_3$), Δk for third harmonic generation (THG) is:

$$\Delta k = 3\omega_1 \{ [v^{MDR}(3\omega_1)]^{-1} - [v^{MDR}(\omega_1)]^{-1} \},$$

where $v^{\text{MDR}}(3\omega_1)$ and $v^{\text{MDR}}(\omega_1)$ are the phase velocities of MDR's at ω_3 and ω_1 , respectively. For the more general case, when $\omega_1 \neq \omega_2 \neq \omega_3$, Δk is only slightly more complicated.

We calculated $v^{\text{MDR}}(\omega_1)$ for MDR's of various mode numbers and mode orders and noted that $v^{\text{MDR}}(\omega_1) \geq c/n(\omega)$, where c is the speed of light in vacuum and $n(\omega)$ is the index of refraction of the liquid at ω . The phase matching of MDR's is greatly improved relative to the phase matching of plane waves propagating in the bulk liquid. Our tentative conclusion is that the TSFG intensity can be selectively increased by tuning a particular MDR to $\omega_1 + \omega_2 + \omega_3$. [Our findings are published in Optics Letters 15, 895 (1990) and the proceedings of a conference in honor of Bloembergen's 70th birthday, see publications #7 and #8, respectively.]

II. Two- and Three-Dimensional Measurements in Flames

The overall objective of our research in this area has been to improve the understanding of combusting flows by the development and application of advanced laser diagnostic techniques to flows of interest. Specific objectives outlined in our original proposal included the following:

1. Measurement of scalar fields and their full three-dimensional gradient within a cross section of a turbulent flame. For these measurements, the flow is simultaneously illuminated with two closely-spaced parallel light sheets. Two cameras are used with each one detecting the image from a single sheet. To ensure that each detector images the scattered light from only one sheet, lasers of different output wavelength or polarization are used and appropriate wavelength-selective or polarizing filters are placed before each camera. After measuring the separation of the two illumination sheets and the spatial resolution of the images from each camera, the gradient vector can be calculated. From a large ensemble of instantaneous

measurements, it will be possible to determine the joint pdf of the scalar and its gradient. This information can be compared directly with current combustion models.

2. Measurement of the temporal evolution of large-scale structures in reacting flows.

Although considerable progress has been made toward increasing the rate at which two-dimensional measurements can be made, the requirements of turbulent flames are quite severe. For many flows of interest, a rate of 5 - 10 kHz is required to follow the evolution of structures. We propose to take advantage of new capabilities of solid state detectors and image intensifiers to attain the framing rate needed to follow the temporal evolution of structures in reacting flows.

3. To refine and extend the three-dimensional measurement techniques already developed. The ability to obtain three-dimensional measurements in turbulent flows has been demonstrated. More work is required, however, to address problems specific to these measurements and to improve their accuracy. The three-dimensional techniques can be expected to go through a phase of refinement and extension to more general experimental configurations similar to that with the two-dimensional measurements.

During the research period, progress was made in all of the proposed areas, along with advances in some areas not foreseen at the time of the original proposal. Specific accomplishments in the development of two- and three- dimensional measurement techniques in flames include the following:

(a) Two-Dimensional Time Evolution in Hydrogen-Air Flames

The fuel-air mixture concentration has been measured as a function of time in the central plane of a turbulent premixed jet flame. A stoichiometric hydrogen-air mixture was uniformly seeded with submicron-sized aerosol particles. The aerosol particles were removed from the flow at the reaction front, and their presence was used to map the concentration of the fuel-air mixture. A sheet of laser light was formed with an Ar⁺ laser and the elastically

scattered light from the flame was recorded in two dimensions at 48 kHz using a framing camera to record 20 frames. The interface between the unburned fuel-air mixture and reaction products was clearly visible in the scattered intensity distribution. From the sequence of images, the motion of the flame front and the convective motion of the gas are evident. The convection velocity of the reacting flow structures was computed from the motions of centroids of unburned gas packets and found to be 90% of the nozzle exit velocity. The burning velocity was also determined by comparing successive realizations in a convecting frame of reference yielding a value ≤ 3 m/s. The effects of seeding were determined to be minimal by comparing the behavior of seeded and unseeded mixtures in a laminar flame. [A report on this work has been published in Combustion Science and Technology, see publication #15.]

The main drawbacks of the technique described above, are the dependence on marker particles to infer the behavior of the flames and the relatively limited image quality available from the framing camera. In an effort to obtain the same information without the limitations imposed by our previous experimental configuration, a second set of experiments was performed. A double pulsed Nd:YAG laser (pulse separation of 100 μ s) was used to illuminate the flow, and Rayleigh scattered light from molecules in the flow was collected by the imaging optics. A high speed rotating mirror displaced subsequent images onto different portions of an intensified CCD detector. Although only a single pair of images was produced, burning and convection velocities can still be obtained, and the quality of the data is considerably improved.

(b) Time Development of Three-Dimensional Structures

The most difficult aspect of obtaining instantaneous three-dimensional data is the requirement that the measurement be made in a time during which the flow is essentially stationary. To relax this constraint, measurements can be performed in forced flows. By causing the flow to evolve in a repeatable fashion, the constraint of making very rapid

measurements is replaced with the condition that the measurement be made at the right phase of the repeatable flow. A further advantage of studying forced flows includes the prospect of utilizing weak scattering mechanisms that would not provide enough signal for single-shot measurements. Since the flow is repeatable, many instantaneous shots can be accumulated to integrate weak signals. Additionally, sequential measurements of several different quantities such as temperature, species, and velocities are also possible.

Another advantage of using forced flows is realized by varying the relative phase of the perturbation and the measurement. In this way, the evolution of the three-dimensional structures can be recorded. A measurement of this type has been performed in our laboratory in an acoustically forced nonreacting jet. The concentration of nozzle fluid was measured at points within a volume using Lorenz-Mie scattering from aerosols seeded into the flow. An animated sequence has been produced that shows the development of a three-dimensional surface of constant concentration. The convection and evolution of the structures is evident. [Our work on three-dimensional measurements was reported at the 22nd Symposium (International) on Combustion as well as in other books and journals, see publications #12, #9, and #10.]

A significant problem related to three-dimensional measurements is the difficulty of representing the large amount of data obtained with these techniques. We have continued to develop new and more efficient means of conveying the information contained in the data. [A discussion of issues involving this aspect of the work was published in a special issue of the journal *Computer* related to visualization in scientific computing, see publication #14.]

Although the data from forced flows are useful in understanding the structure and evolution of the early stages of a turbulent flow, forced flows cannot be expected to be an accurate representation of fully developed turbulence. Therefore, we also developed a technique for recording a pair of three-dimensional data sets separated in time by a short (100 μ s) interval. The technique is based on the use of a series of closely spaced parallel laser illumination sheets of different wavelengths. A single CCD detector is used with color

filters and image displacing wedges which cause the elastic scattering from each illumination sheet to be imaged onto a separate region of the detector. To obtain temporal information, a high speed rotating mirror is again used to displace the images onto different regions of the detector.

In our initial experiment, four laser sheets were formed from the second-harmonic output of a Nd:YAG laser and from the first-, second-, and third-order stimulated Raman scattering from a liquid filled cell pumped by a portion of the 532 nm beam. The laser was double pulsed, and the Lorenz-Mie scattering from an aerosol-seeded premixed flame was recorded at two times separated by an interval of 100 μ s (i.e., a total of eight images is recorded on the CCD detector). This data allow investigation of the effect of curvature on the burning velocity. [This work is described in publication #19.]

(c) Supersonic Visualization

In a series of experiments performed in collaboration with Robert Dibble of Sandia Laboratories and Godfrey Mungal of Stanford, a simple Rayleigh-scattering technique was developed for visualizing mixing and shock structure in supersonic flows. The elastically scattered light from a thin laser sheet intersecting an underexpanded air-into-air jet was imaged onto a CCD detector. A small amount of humidity in both the nozzle and ambient air resulted in the formation of very small condensed-phase water droplets. The resulting digital images clearly showed the Mach disk and triple-point shock structures as well as large-scale structures near the edge of the jet and along the slip discontinuity. The ability to visualize regions of rapid mixing in this simple way should be a valuable tool for studying supersonic mixing.

The relationship of the signal intensity to the flow properties is complex since the temperature and pressure of the gas vary as do the number density and size distribution of the particles. However, the large dynamic range provided by the CCD detector allows some information on the nature of the condensed-phase particles to be obtained. An investigation

of the pixel-to-pixel signal and noise characteristics within a small area of the flow containing particles (in which the particle size and number density can be expected to be constant) revealed that (1) the signal is larger than in regions not containing particles by as much as a factor of 100 and (2) the signal/noise ratio is much smaller than in regions containing no particles. We have used this information in several ways to estimate the characteristics of the particles formed. First, the number of condensed-phase scatterers was determined by considering the noise statistics of particle-containing regions. Second, the magnitude of the signal was used along with the expression for the Rayleigh scattering cross section as a function of particle radius to provide an estimate of the size of the scattering particles. We estimate that the largest of the particles have a radius of 30 nm and, therefore, have very little slip relative to the gas flow, even at supersonic velocities. Finally, by considering the total number of water molecules present in the flow before condensation, a check on the validity of the results of particle size and number density is obtained as well as some insight into the condensation process responsible for forming particles. [One of the images from this experiment won an award in the annual Picture Gallery sponsored by the Division of Fluid Dynamics of the American Physical Society and appeared in *Physics of Fluids*, see publication #17].

(d) Scalar Gradient Measurements

Some of the results obtained in the work on measuring the full three-dimensional scalar gradient have been used by other researchers to advance theories of turbulent nonpremixed combustion. Most notably, Prof. Robert Bilger at the University of Sydney has used our data to propose a new model for nonpremixed combustion based on quasi-equilibrium distributed reaction (QEDR) zones. The use of the unique data attainable only through advanced diagnostic techniques as an input to combustion modeling is a positive step. [Some of the results of this collaborative effort have been published, see publication #13].

(e) Simultaneous CH and CH₄ Mapping

Another set of collaborative experiments was done with researchers at Sandia National Laboratories in Livermore, California. In this work, a technique was developed for simultaneously monitoring the concentration of CH₄ via Raman scattering and CH via fluorescence has been developed. The experimental configuration for this experiment is similar to that used for the scalar gradient measurements. In this case, however, two overlapping laser sheets are used and each of the two detectors monitors a different light-scattering mechanism. The simultaneous mapping of the fuel concentration (CH₄) and the location of the flame front (marked by the CH) give insight into the relationship between fuel-air mixing and subsequent combustion. [The results of these experiments have been reported, see publications #11 and #18].

(f) Investigation of Differential Diffusion Effects

One of the aspects of reacting flows that makes their complete characterization so difficult is the large number of species present. Even a simple flame contains fuel, oxidizer, intermediates, combustion products, and relatively inert components. In general, each of these components will possess a unique diffusion coefficient. In most of the work in turbulent combustion, however, the simplifying assumption is made that a single diffusion coefficient can be used to characterize all of the species present and that differential diffusion effects can be neglected at reasonably high Reynolds numbers.

A Rayleigh scattering experiment was performed that allowed some of the simplifying assumptions to be checked by providing a direct measurement of differential diffusion effects. In the experiment, hydrogen (which has a Rayleigh cross section lower than that of air) and Freon (which has a higher Rayleigh cross section) were mixed in a ratio so that the effective Rayleigh cross section of the mixture is precisely the same as that of air. The mixture exited into a slowly coflowing stream of air through a cylindrical nozzle, and the

resulting turbulent jet was investigated using planar Rayleigh scattering. Because the Rayleigh cross section of the mixture was the same as that of the air, the Rayleigh signal is independent of the mixing of the H₂/Freon jet with the air. However, if the ratio of H₂ to Freon changes due to the different diffusivities of the components, the Rayleigh signal will vary, with Freon rich regions providing more Rayleigh scattering and H₂ rich regions scattering less than the air or regions of the correct ratio.

The experiment showed that the effect is readily observable and that the magnitude of the effect is larger than that predicted by previous modeling efforts. Measurable differential diffusion is found at Reynolds numbers as high as 20,000 and as far downstream as 30 nozzle diameters. In addition to H₂/Freon/air experiments, differential diffusion is also observed when a mixture of H₂ and CH₄ is issued from the nozzle. (H₂ and CH₄ have less disparate diffusion coefficients than H₂ and Freon and may have more implications for combustion work.)

Although the experiment to measure these effects was proposed nearly ten years ago, previous attempts to make the measurement using single-point techniques were not successful. Two critical aspects in the success of the current experiment were (1) the high sensitivity imaging capability of the CCD detector and (2) the use of a second CCD detector to ensure that the Rayleigh cross section of the H₂/Freon mixture was precisely the same as that of air. [The results of the experiments have implications for both theorists and experimentalists, see publication #16].

PUBLICATIONS RESULTING FROM THE RESEARCH

I. Nonlinear Spectroscopy of Multicomponent Droplets

1. J.-B. Zheng, W.-F. Hsieh, S.-C. Chen, and R.K. Chang, "Temporally and Spatially Resolved Spectroscopy of Laser-Induced Plasma from a Droplet," *Opt. Lett.* **13**, 559 (1988).
2. J.-Z. Zhang and R.K. Chang, "Shape Distortion of a Single Water Droplet by Laser-Induced Electrostriction," *Opt. Lett.* **13**, 916 (1988).
3. J.-B. Zheng, W.-F. Hsieh, S.-C. Chen, and R.K. Chang, "Growth, Decay, and Quenching of Stimulated Raman Scattering in Transparent Liquid Droplets," in Laser Materials and Laser Spectroscopy, Z. Wang and Z. Zhang, eds. (World Scientific, Singapore, 1989), p. 259.
4. J.-Z. Zhang, G. Chen, and R.K. Chang, "Pumping of Stimulated Raman Scattering by Stimulated Brillouin Scattering within a Single Droplet: Input Laser Linewidth Effects," *J. Opt. Soc. Am. B* **7**, 108 (1990).
5. R.K. Chang and A.S. Kwok, "High Intensity Laser Beam Interactions with Single Droplets," in Proceedings of the AGARD Conference on Atmospheric Propagation in the UV, Visible, IR and MM-Wave Region and Related Systems Aspects, Copenhagen, October 9-13, 1989, AGARD CP-454 (NATO, Neuilly Sur Seine, France, 1990), p. 20-1.
6. A.S. Kwok, C.F. Wood, and R.K. Chang, "Fluorescence Imaging of CO₂ Laser-Heated Droplets," *Opt. Lett.* **15**, 664 (1990).
7. D.H. Leach, W.P. Acker, and R.K. Chang, "Effect of the Phase Velocity and Spatial Overlap of Spherical Resonances on Sum-Frequency Generation in Droplets," *Opt. Lett.* **15**, 895 (1990).
8. R.K. Chang, "Some of Bloembergen's Nonlinear Optical Effects Revisited in Single Micrometer-Sized Droplets," in Resonances, M.D. Levenson, E. Mazur, P.S. Pershan, and Y.R. Shen, eds. (World Scientific, Singapore, 1990), p. 200.

II. Two- and Three-Dimensional Measurements in Flames

9. M.B. Long, B. Yip, M. Winter, and J.K. Lam, "Measurement of the Topology of Large-Scale Structures in Turbulent Reacting Flows," in *Turbulent Reactive Flows*, R. Borghi and S. N. B. Murthy, eds. (Springer-Verlag, New York, 1989), p. 1.
10. B. Yip, R.L. Schmitt, M.B. Long, "Instantaneous Three-Dimensional Concentration Measurements in Turbulent Jets and Flames," *Opt. Lett.* **13**, 96 (1988).

11. M. Namazian, R.L. Schmitt, and M.B. Long, "Two-Wavelength Single Laser CH and CH₄ Imaging in a Lifted Turbulent Diffusion Flame," *Appl. Opt.* **27**, 3597 (1988).
12. M.B. Long and B. Yip, "Measurement of Three-Dimensional Concentrations in Turbulent Jets and Flames," *Proceedings of the Twenty-Second Symposium (International) on Combustion*, (The Combustion Institute, Pittsburgh, 1988), p. 701.
13. R.W. Bilger, B. Yip, M.B. Long, and A.R. Masri, "An Atlas of QEDR Flame Structures," *Combust. Sci. Tech.* **72**,137 (1990).
14. M.B. Long, K. Lyons, and J.K. Lam, "Acquisition and Representation of Two- and Three-Dimensional Data from Turbulent Flows and Flames," *Computer* **22**(8), 39 (1989).
15. M. Winter and M.B. Long, "Two-Dimensional Measurements of the Time Development of a Turbulent Premixed Flame," *Combust. Sci. Tech.* **66**, 181 (1989).
16. A.R. Kerstein, R.W. Dibble, M.B. Long, B. Yip, and K. Lyons, "Measurement and Computation of Differential Molecular Diffusion in a Turbulent Jet," *Proceedings of the Turbulent Shear Flows Conference*, August 1989, Stanford, CA.
17. B. Yip, K. Lyons, M. Long, M.G. Mungal, R. Barlow, and R. Dibble, "Visualization of a Supersonic Underexpanded Jet by Planar Rayleigh Scattering," *Phys. Fluids A*, **1**(9), 1449 (1989).
18. R.S. Barlow, R.W. Dibble, S.H. Stårner, R.W. Bilger, D.C. Fourquette, and M.B. Long, "Reaction Zone Structure in Dilute Methane Jet Flames Near Extinction," 28th Aerospace Sciences Meeting, January 1990, AIAA-90-0732.
19. J. Frank, K. Lyons, and M.B. Long, "Technique for Three-Dimensional Measurements of the Time Development of Turbulent Flames," *Opt. Lett.*, (in press).

SCIENTIFIC COLLABORATORS

In addition to the Co-Principal Investigators, the following people have participated in this project:

I. Nonlinear Spectroscopy of Multicomponent Droplets

Postdoctoral Associates: William P. Acker
Shu-Chi Chen (Hong Kong Baptist College)
Carol Wood
Jia-Biao Zheng (Fudan University)

Graduate Students: Gang Chen
Alfred Kwok
Wen-Feng Hsieh
Jian-Zhi Zhang

II. Two- and Three-Dimensional Measurements in Flames

Postdoctoral Associates: Brandon Yip
Ruichen Jin

Graduate Students: Michael Winter
Brandon Yip
Kevin Lyons
Sumit Sen
Jonathan Frank

LECTURES PRESENTED ABOUT THE RESEARCH

Richard K. Chang:

"Nonlinear Optics in Single Droplets," Physics Activity Club, Yale University, New Haven, CT, April 8, 1988.

"Nonlinear Optical Spectroscopy of Single Droplets," AFOSR Contractors' Review Meeting, California Institute of Technology, Pasadena, CA, June 14, 1988.

"Four-Wave Mixing in Single Droplets," XIII International Conference on Coherent and Nonlinear Optics, Minsk, USSR, September 6-9, 1988 (invited talk).

"Nonlinear Optics from a Single Micrometer-Size Liquid Droplet," Center of Lightwave Technology, University of Toronto, Toronto, Ontario, Canada, November 18, 1988.

"Laser-Induced Breakdown and Shape Distortions in Transparent or Absorbing Liquid Droplets" and "Nonlinear Optical Effects in a Single Liquid Droplet: Third-Order Sum Frequency Generation and Stimulated Brillouin Scattering," 5th Annual Workshop on the Physics of Directed Energy Propagation in the Atmosphere, New Mexico State University, Las Cruces, NM, February 28-March 1, 1989.

"Nonlinear Optical Spectroscopy from Liquid Droplets," Conference on lasers and Electro-Optics (CLEO), Baltimore, MD, April 27, 1989 (invited talk).

"Nonlinear Optical Interactions in Droplets," Mechanical Engineering and Physics Department of the University of Nebraska, Lincoln, NE, May 5, 1989.

"Nonlinear Optical Scattering of Single Liquid Droplets," Physics Department of Iowa State University, Ames, IA, May 8, 1989.

"High Intensity Laser Beam Interactions with Single Droplets," (Invited) AGARD Conference on Atmospheric Propagation in the UV, Visible, IR and MM-Wave Region and Related Systems Aspects, Copenhagen, October 9-13, 1989.

"Nonlinear Optical Effects in Single Liquid Droplets," Department of Physics, University of Texas at Dallas, October 25, 1989.

"Nonlinear Optical Effects in Single Liquid Droplets," Columbia Radiation Laboratory, Columbia University, November 20, 1989.

"Third-Order Nonlinear Effects in Droplets," University of Connecticut, Department of Chemistry, Storrs, Connecticut, January 31, 1990.

"The Droplet as an Optical Cavity," Department of Physics, Yale University, February 21, 1990.

"Enhancement of the Third-Order Sum-Frequency Generation in Droplets: Improved Phase Matching of Morphology-Dependent Resonances," Sixth Annual Workshop on the Physics of Directed Energy Propagation in the Atmosphere," Las Cruces, New Mexico, March 6-7, 1990.

"Some of Bloembergen's Nonlinear Optical Effects Revisited in a Micrometer-Sized Droplet," (Invited) Symposium in Honor of Prof. N. Bloembergen's 70th Birthday, Harvard University, May 27, 1990.

"Nonlinear Spectroscopy of Multicomponent Droplets," AFOSR Contractors Meeting, Atlanta, Georgia, June 14-15, 1990.

"Coherent Emission Processes in Single Droplets," United Technologies Research Center, East Hartford, Connecticut, June 22, 1990.

"Stimulated Anti-Stokes Raman Scattering from Monodispersed Droplets," 1990 CRDEC Scientific Conference on Obscuration and Aerosol Research," Aberdeen Proving Ground, Maryland, June 25-28, 1990.

"Applications of Nonlinear Spectroscopy to Droplet Streams and Sprays," Air Force Aero-Propulsion Laboratory, Wright Patterson AFB, OH, July 10, 1990.

Marshall B. Long:

"Two- and Three- Dimensional Measurements in Flames," AFOSR Contractors Meeting on Rocket Propulsion, Monrovia, California June 14-15, 1988.

"Measurement of Three-Dimensional Concentrations in Turbulent Jets and Flames," Twenty-Second Symposium (International) on Combustion, University of Washington, Seattle, Washington, August 14-19, 1988.

"Laser Imaging Techniques in Turbulent Flames," Department of Mechanical Engineering Seminar Series, Cornell University, Ithaca, New York, September, 1988.

"Differential Diffusion in Jets," 12th Meeting of the Sandia Cooperative Group on the Aerothermochemistry of Turbulent Combustion, GE Corporate R&D Center, Schenectady, New York, October 10-11, 1988.

"Multi Dimensional Imaging in Gaseous Flows," HTGL Seminar, Stanford University, Palo Alto, California, March 15 1989.

"Multi-Dimensional Measurements in Turbulent Flows and Flames," University of Connecticut , Mechanical Engineering Department Spring Seminars, Storrs, Connecticut, April 3, 1989.

"Multidimensional Imaging," Gordon Research Conference on the Chemistry and Physics of Laser Diagnostics, Plymouth, New Hampshire, July 17-21, 1989.

"Advanced Imaging Techniques for Combustion Research," Combustion Dynamics Workshop/Review, Lawrence Berkeley Laboratory, Berkeley, California, October 16-17, 1989.

"Scalar Measurements in Two, Three and Four Dimensions," (Invited) 8th International Congress on Applications of Lasers and Electro-Optics (ICALEO), Orlando, Florida, October 16-18, 1989.

"Imaging Diagnostics Update," 14th Meeting of the Sandia Cooperative Group on the Aerothermochemistry of Turbulent Combustion, Nashville, Tennessee, November 6-7, 1989.

"Multidimensional Measurements in Turbulent Flows and Flames," University of Sydney, Australia, February 1990.

"Digital Multidimensional Imaging and its Interpretation," (Invited), NATO Advanced Study Institute on Combusting Flow Diagnostics, Algarve, Portugal, April 15-29, 1990.

"Two- and Three-Dimensional Measurements in Flames," AFOSR/ONR Contractors Review Meeting on Propulsion, Atlanta, Georgia, June 14-15, 1990.

"Multi-Dimensional Laser Diagnostics for Turbulent Flows," US-USSR Workshop on Interactive Thermo-Combustion Effects in Turbulent Mixing, Novosibirsk, USSR, August 20-25, 1990.

"Laser Diagnostics for Flame Structure Determination." Mechanism of Non-uniform Combustion Conference, Tokyo, Japan, September 10-11, 1990.

"Advances in Quantitative Imaging Diagnostics of Reacting Flows," OSA Annual Meeting, Boston, Massachusetts, November 4-10, 1990.

"2-D Images of Water Condensation in Supersonic Flows," ICALEO Meeting, Boston, Massachusetts, November 4-10, 1990.

"Multi-Dimensional Laser Diagnostics for Flame Studies," Eastern Section Meeting of the Combustion Institute, Orlando, Florida, December 3-5, 1990.

INTERACTIONS WITH OTHER LABORATORIES

Richard K. Chang:

Atmospheric Science Laboratory, White Sands Missile Base, NM
Dr. R. Pinnick and Dr. D. Pendleton

Ballistic Research Laboratories, Edgewood, MD
Dr. Richard Beyer and Dr. A Miziolek

CRDEC, Aberdeen Proving Ground, MD
Dr. J. White and Dr. I. Sindoni

Sandia National Laboratories, Livermore, CA
Dr. Peter Mattern, Dr. David Dandy, Dr. Larry Rahn, and Dr. Roger Farrow

Texaco Research Center, Beacon, NY
Dr. Rodney Sung and Dr. William Acker

United Technology Research Center, East Hartford, CT
Dr. Alan Eckbreth, Dr. Greg Dobbs, and Dr. Larry Boedecker

Air Force Aero-Propulsion Laboratory, Wright Patterson AFB, OH
Dr. Mel Roquemore and Dr. Tom Jackson

Marshall B. Long:

Sandia National Laboratories, Livermore, CA
Dr. Robert Dibble, Dr. Rob Barlow, Dr. Alan Kerstein

Texaco Research Center, Beacon, NY
Dr. William Acker

University of Sydney, Sydney, NSW, Australia
Prof. Robert W. Bilger, Dr. Sten Stårner

Altex Technologies Corp., Los Gatos, CA
Dr. Mehdi Namazian

Stanford University, Palo Alto, CA
Prof. Godfrey Mungal