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Significant progress has been made in the following areas: (I) Nonlinear spectr... of micrometer sized droplets, and (II) Development and application of two- and three-dimensional scalar and velocity measurement techniques in flames.

In the nonlinear spectroscopy area, the following achievements are reported: (1) Fluorescence seeding of the stimulated Raman scattering (SRS) in order to increase the detectivity (by as much as 10X) of minor species in a multicomponent liquid droplet that contains a fluorescent dye; (2) Temporal precession of the SRS as a means of determining the shape distortion (7 parts out of 10³) of a flowing droplet because of inertial force; (3) Preliminary measurements of the relative evaporation rates of 15 droplets in a segmented stream, where the few lead droplets evaporate faster than the remaining droplets; and (4) Suppression of lasing by the SRS and amplification of the SRS in microdroplets, leading to an increased SRS signal.

Achievements in multi-dimensional scalar and velocity imaging includes the following: (1) Combined two-dimensional images of the Rayleigh scattering and acetaldehyde fluorescence provided new information of the mixture fraction of a turbulent nonpremixed acetaldehyde flame; (2) Tomography for three-dimensional measurements in a flame, where five CCD cameras were used to measure the flame luminosity of a methane diffusion flame; and (3) Simultaneous particle image velocimetry (PIV) and CH fluorescence measurements (preliminary) were made in order to determine the velocity field and a scalar quantity at the flame front of a premixed flame.

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

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February 1, 1992 - January 31, 1993

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INTRODUCTION

During the second year 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 and velocity mapping in flames. Specific details of our accomplishments regarding nonlinear interactions in droplets and multi-dimensional 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

Nonlinear optical spectroscopy has the potential of determining the chemical species and physical properties of multi-component liquid droplets in a spray combustor. Our research is directed toward the following two goals: (1) furthering the understanding of nonlinear optical processes within individual droplets; and (2) extending the application of nonlinear optical techniques for chemical identification of multicomponent fuel droplets and physical characterization (e.g., the evaporation rate and shape distortions) of flowing and interacting droplets. The spherical liquid-air interface of a micrometer-sized droplet has the following unique properties: (a) refracts and reflects the incoming radiation, causing an increase of the internal intensity of the incident radiation; (b) enhances the spontaneous emission rates and, hence, increases the lasing, Brillouin, and Raman gain coefficients; and (c) provides optical feedback (with high Q) for the internally generated lasing, stimulated Brillouin, and stimulated Raman radiation.

Since the previous AFOSR Annual Report, five papers that were either "submitted" or "to be published" have now appeared in print (see publications #1 through #5 on page 15). Under preparation is a chapter of a book associated with the summer school lecture

R.K. Chang delivered in Aalborg, Denmark, August, 1992. The intent of this chapter is to review the field of nonlinear optical processes in droplets since 1990 when R.K. Chang wrote a review entitled, "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.

The main research results during the past year are: (1) fluorescence seeding of stimulated Raman scattering as a means of increasing the weaker-gain Raman signal; (2) precession of stimulated Raman scattering as a means of determining the droplet shape distortion; (3) determination of relative evaporation rates of a segmented stream of interacting droplets; and (4) suppression of lasing by stimulated Raman scattering in droplets. The following is a brief summary of the research accomplishments in four areas of nonlinear optical interactions in droplets and their applications to droplet diagnostics.

(1) Fluorescence Seeding of Stimulated Raman Scattering (SRS)

Conventionally, SRS builds up from spontaneous Raman noise. In the absence of external seeding by injecting radiation at the Stokes wavelength, the SRS processes can be considered as the amplification of spontaneous Raman noise by e^{30} . However, when the droplet contains dye molecules that fluoresce within the Stokes Raman wavelength region, the Stokes SRS no longer needs to start only from spontaneous Raman noise but can also be initiated from fluorescence within the Raman linewidth. Even though the typical dye concentration is extremely low (e.g., 10^{-7} to 10^{-6} M), the fluorescence noise is still much larger than the spontaneous Raman noise, because the fluorescence cross section ($\sigma \approx 10^{-16}$ cm²) is many orders of magnitude larger than the spontaneous Raman cross section ($\sigma \approx 10^{-30}$ cm²). Fluorescence seeding effectively lowers the input-pump laser intensity needed to achieve a certain detectable SRS intensity. The SRS intensity is allowed to build up from a much larger initial value, that is determined by the fluorescence of the dye molecules rather than from the weaker spontaneous Raman intensity. Moreover, fluorescence seeding

fluorescence seeding also increases the growth rate of the SRS intensity, because the parametric growth rate is dependent on the product of the input-pump intensity and the Stokes intensity that has contributions from both the Raman and fluorescence emissions.

For species detection of a multi-component fluid droplet by SRS detection, fluorescence seeding can preferentially increase the SRS intensity of the minor species. The fluorescence spectrum is independent of the pump wavelength. By contrast, the Raman spectrum is dependent on the pump wavelength, because the Stokes Raman frequency shift is always equal to the vibrational frequency of the molecule. Good spectral overlap between the fluorescence and the Raman scattering of the minor species is the requirement for selective fluorescence seeding of the SRS of the minor species. In addition, it is required that the Raman scattering of the major species must be beyond the fluorescence emission spectrum. Although we have not demonstrated that we can achieve the improved sensitivity of SRS of minor species, we have demonstrated the "proof of principle" by improving the SRS sensitivity of a weaker-gain Raman mode of ethanol. For ethanol droplets containing 10^{-7} M Rhodamine 6G dye, the SRS intensity of the weaker gain C-C-O at 882 cm^{-1} is compatible with the SRS intensity of the stronger gain C-H mode at 2928 cm^{-1} . For more details of our preliminary results see publication #6 on page 15. Further research is needed before selective fluorescence seeding of Raman modes associated with minor species can be exploited and provide quantitative species concentration information.

(2) Precession of the SRS as a Means of Determining Droplet Shape Distortions

In the previous AFOSR Annual Report, we reported on the observation of frequency splitting of degenerate spherical cavity mode by using a Fabry-Perot interferometer to analyze spectrally the SRS emerging from a single droplet that is flowing in a continuous stream of droplets. Because of the inertial effect, the droplet shape is distorted into a prolate spheroid, with the axis of symmetry (z-axis containing the droplet

poles) along the flow direction. Unlike a sphere where all the great circles have the same circumference, the great ellipses of a prolate spheroid do not have the same length. Consequently, the round trip distance is slightly larger around the droplet equator and hence, the morphology-dependent resonance (MDR) of the droplet at the equator shifts to a longer wavelength. Furthermore, the round trip distance is slightly smaller around the droplet poles and hence, the MDR of the droplet at the poles shifts to a shorter wavelength. The wavelength positions of the MDRs with different inclination angle θ from the droplet poles are explicitly given from the perturbation theory developed by the group at the Chinese University of Hong Kong. In between the equator and the pole, the MDRs appear at discrete wavelengths. We have deduced the droplet shape distortion to be one part in 10^3 by fitting the observed frequency shifts of MDRs at different θ (or frequency splittings among the MDRs). For details see a review of our frequency splitting results in publication #8 on page 15.

During this year, we have observed that the SRS (on a MDR inclined at θ) will precess in time. From the ray-optics view point, the radiation that circulates once around the oblate droplet rim or around a great ellipse does not begin and end at the same point. The end point is shifted by a small angle $d\phi$ because of the shape distortion. After many trips around the droplet or after one precession period, the radiation will return to an inclination of θ . With a streak camera, we were able to measure the precession period of the SRS that is on a MDR inclined at angle θ . By using the perturbation theory for MDR shifts or splittings at a function of θ , with the help of Professor Kenneth Young of the Chinese University of Hong Kong, we were able to deduce the droplet shape distortion from the precession period or frequency. The droplet shape distortion was deduced to be 7 parts out of 10^3 . The discrepancy between the previous frequency splitting measurement (of 1 part out of 10^3) and the present precession time measurement (of 7 parts out of 10^3) was attributed to the following experimental parameters: (1) the droplets were larger for the time-dependent experiment; and (2) the droplets were traveling faster in the time-dependent

experiment. Because the shape distortion is highly dependent on both the Weber and Reynolds numbers, we were satisfied that the time measurements gave a larger distortion than the frequency measurements. For details of the precession of the SRS inclined at θ , see publications #7 and #8 on page 15.

(3) Relative Evaporation of Droplets in a Segmented Stream

In 1984 we used the frequency shift in the lasing spectrum from individual droplets in a continuous stream to determine the evaporation of droplets [for details see H.-M. Tzeng, K. F. Wall, M. B. Long, and R. K. Chang, "Evaporation and Condensation Rates of Liquid Droplets Deduced from Structure Resonances in the Fluorescence Spectra," *Opt. Lett.* 9, 273 (1984)]. Since then, we were inspired by the theoretical papers of Prof. William Sirignano on three interacting droplets that exhibit different evaporation rates. In the calculation, the lead droplet has the largest evaporation rate and the third droplet has the evaporation rate of a continuous stream with the same droplet-droplet separation.

During this year, we initiated an experiment to measure the evaporation rate of closely spaced droplets in a segmented stream. A pair of constant-voltage electrostatic-deflection plates was used to deflect the charged droplets in an undesired segment. The uncharged droplets in the desired segment will remain undeflected. Because of induced charges, the lead droplet of the desired segment always appeared to be $(2)^{1/3}$ or $(3)^{1/3}$ times larger than the trailing droplets. We are currently using a charge compensation technique (developed for ink-jet printers) to attempt to produce a lead droplet of equal radius with respect to the trailing droplets.

In spite of the fact that our lead droplet is larger than the trailing droplets, we noticed several interesting features regarding the evaporation rates of individual droplets: (1) the lead droplet evaporates faster than all the trailing droplets; (2) each droplet evaporates a little faster than its trailing droplet; and (3) only by the 6th trailing droplet, does the evaporation rate equal the evaporation rate of a continuous stream.

By using the same apparatus, we were amazed that with the spectral resolution of a high-resolution spectrograph (not an interferometer) we can resolve the frequency shifts of a deformed droplet associated with the inertial effect. In the future, we will be able to determine **simultaneously** both the droplet evaporation rate and the inertial force-induced droplet shape distortion for each droplet in the segmented stream. Furthermore, since our spectrograph preserves the spatial information, we will be able to measure the change in the droplet-droplet separation at two different times, deducing the relative drag coefficient for each droplet in the segmented stream. We are actively pursuing this experiment, because of the obvious implication our measurement will have on verifying the hydrodynamic computer codes for fuel sprays dynamics. Our main challenge is how to make the lead droplet have the same volume as the trailing droplets. For details see publication #9 on page 15.

(4) Suppression of Lasing by Stimulated Raman Scattering in Microdroplets

In an attempt to further our understanding of fluorescence seeding of SRS by the dye molecules contained in the fuel droplets, we varied the dye molecule concentration from 10^{-8} to 10^{-3} M. For dye concentration above 10^{-6} M, lasing by the dye molecules is inevitable at the pump intensity necessary to observe SRS gain. Consequently, we were led into studying the competition between two interactions: (1) lasing of the dye molecules; and (2) SRS of the high gain vibrational modes associated with the host liquid molecules.

We noticed that with very lower pump intensity, the emission spectrum from the dye-doped droplets was dominated by the laser emission. However, when the pump intensity is increased, the emission spectrum is dominated by high gain SRS peak. To our surprise, with increased pump intensity, the lasing intensity at wavelengths away from the dominant SRS peak actually decreased in strength. The suppression of lasing and the extra amplification of the SRS by the inverted dye population are two novel features that require further investigation. To date, we understand that the lasing gain per round trip is pinned

to a value to be just above the round trip loss at the lasing wavelength. However, the SRS gain is not pinned and can continue to grow until intensity depletion saturates the SRS intensity. The Raman photons can stimulate transition from the upper laser state to the lower laser state, depleting the population inversion for lasing at wavelengths outside the Raman peak. Consequently, the overall gain of the Raman photons is increased by the homogeneously broadened dye-molecule gain profile. However, the lasing gain of the dye molecules is decreased by the pump-intensity dependent Raman gain. After we reach a deeper understanding of these competing gain processes, we will then determine whether or not we can exploit the extra lasing gain, in addition to the fluorescence seeding, in order to increase the SRS intensity from trace molecules in the droplet. For more details of our preliminary findings on the suppression of lasing by SRS, see publication #10 on page 15.

II. Two- and Three-Dimensional Measurements in Flames

Despite considerable study and some notable progress, questions persist as to the structure of turbulent nonpremixed flames. For example, information on the relative scales of the flame and the turbulence is needed to better understand the interaction between the turbulent flow field and the combustion chemistry. Other questions relate to the applicability of flamelet concepts for characterizing turbulent flames and to the importance of the flame structure in determining pollutant formation. Better understanding of these aspects of the flame structure is an important step in improving models of turbulent nonpremixed flames. Laser-based imaging diagnostics are particularly well suited for providing structural information in turbulent nonpremixed flames. The high-resolution multi-dimensional measurements of scalars provided by these measurements are critical to understanding both fundamental phenomena in flames and the operating characteristics of practical devices. In much of the work done to date, however, the scalar quantity measured has been dictated more by the capabilities of the diagnostic technique than by the need of

combustion modelers or those using the diagnostics for testing in development of real combustion.

Within the past year of AFOSR funding, we have initiated a set of experiments in which we hope to develop a new imaging technique for determining the mixture fraction; a quantity directly relevant to modeling nonpremixed turbulent combustion. The availability of mixture-fraction images in flames will be an important step in resolving issues related to the scales of flame structure and the operative burning regimes within turbulent nonpremixed flames.

(1) Two-Dimensional Measurements of the Mixture Fraction

The mixture fraction (defined as the mass fraction of atoms that originated from the fuel stream) is of critical importance for modelers of turbulent nonpremixed combustion. Since this conserved scalar quantity is not affected by the chemical reaction, it gives valuable information on the mixing process. Under certain conditions, (high Damkohler number, chemical equilibrium, equal diffusivities) this conserved scalar can be used to derive essentially all of the quantities of interest in the flame. Experimentally, the determination of the mixture fraction in nonpremixed flames is quite difficult since it involves simultaneous monitoring of a large number of species. At Sandia Laboratory the mixture fraction has been determined at a single point from measurements of all major species using spontaneous Raman scattering. Results from these studies are among the most complete sets of experimental data available in turbulent flames. The data from single-point measurements are incomplete, however, because of the lack of multi-dimensional information required to obtain gradients. The scalar dissipation, χ (defined as $\chi = 2D \nabla \xi \cdot \nabla \xi$ with D the diffusivity), determines the rate of molecular mixing and is widely used in modeling turbulent reacting flows. Therefore, techniques capable of two-dimensional or preferably three-dimensional measurements of the mixture fraction are needed.

In nonreacting flows, determination of the mixture fraction is straightforward since only a single species must be monitored and quantitative information in two- and three dimensions is available. In order to investigate issues of flame structure, Prof. R. Bilger used cold flow images of the mixture fraction recorded in our laboratory to infer flame structures in a turbulent nonpremixed H₂ flame. [For more information see R. W. Bilger, "The Structure of Turbulent Nonpremixed Flames," Twenty-Second Symposium (Int'l) on Combustion, The Combustion Institute, (1989) p.475 and R. W. Bilger, B. Yip, M. B. Long, and A. R. Masri, "An Atlas of QEDR Flame Structures," Combust. Sci. Tech. 72, 137 (1990)]. Because image data were available, it was possible to calculate the scalar dissipation as well as reaction rates in this zero heat release "flame". The use of cold flow images to infer nonpremixed flame structure is also used in a recent study by Dahm et al. [For more detail see K. A. Buch, W. J. A. Dahm, R. W. Dibble, and R. S. Barlow, "Structure of Equilibrium Reaction Rate Fields in Turbulent Jet Diffusion Flames," accepted for publication in Twenty-Fourth Symposium (Int'l) on Combustion, Sydney, Australia, July 5-10, 1992]. In all of these studies, valuable insight is gained from the experimental images. However, since the images that served as a basis for the analysis were from cold flows, the heat release is not accounted for and questions persist as to the applicability of the conclusions to real flame.

One of the attractive aspects of mixture fraction imaging is the potential to calculate the scalar dissipation, which involves the square of the mixture fraction gradient. However, the scalar dissipation results are extremely sensitive to noise in the experimental data. It is critical to obtain images with high signal-to-noise ratio. Because of the weakness of the Raman process, prospects for extending the single-point Raman measurement of the mixture fraction to two dimensions are remote. A means of determining the mixture fraction without measuring all of the major species has recently been proposed by Stårner et al. [More detail is available in S. H. Stårner, R. W. Bilger, R. W. Dibble, and R. S. Barlow, "Measurements of Conserved Scalars in Turbulent

Diffusion Flames," *Combust. Sci. Tech.* **86**, (1992) p.223]. In this work it was shown that the mixture fraction can be determined in three different ways from simultaneous measurement of only two quantities. The validity of the method requires a unity Lewis number and a one-step reaction between fuel and oxidizer. The measured quantities are used to form a conserved scalar from which the mixture fraction is determined in an iterative process. During the past year we performed a set of preliminary experiments in which the mixture fraction was obtained from images of the Rayleigh scattering and the fuel concentration. These experiments were conducted in our laboratory with the collaboration of researchers at the University of Sydney.

The flame selected in this study was a turbulent nonpremixed acetaldehyde flame. Acetaldehyde (CH_3CHO) was chosen for its relatively high fluorescence yield and small variation of fluorescence intensity with temperature, which allow the fuel concentration to be found directly from the acetaldehyde fluorescence. Another attractive characteristic relates to the flame chemistry of acetaldehyde, which has been shown to be quite close to that of methane. An alternative means of measuring fuel concentration is Raman imaging, but because of the need for high signal-to-noise ratio to determine scalar dissipation, the more efficient fluorescence process was chosen.

In our initial Rayleigh scattering/acetaldehyde fluorescence imaging experiments, the flow was illuminated by two overlapping laser sheets. The first, formed from the second harmonic of a Nd:YAG pumped dye laser (320 nm, 10 mJ per 10 ns pulse, 10 mm sheet height), excited fluorescence from the acetaldehyde fuel. The second illumination sheet, from a flashlamp-pumped dye laser (532 nm, 2 J per 2 μs pulse, 2 cm sheet height), excited Rayleigh scattering. The fuel fluorescence was imaged onto a gated-image intensifier that was optically coupled to a cooled CCD detector. The Rayleigh scattering was imaged onto a separately gated intensifier optically coupled to the second CCD detector. The two lasers and image intensifiers were fired sequentially with a pulse separation of $\sim 3 \mu\text{sec}$. This temporal separation ensured that there was no interference

between the detected signals from the two laser sheets, but was short compared to the smallest flow time scales.

The raw images were corrected for background and non uniform detector response. In addition, since the magnification of the two images and the illumination sheet size were different, the necessary rotation, translation, scaling, and cropping were applied to the images to allow them to be compared on a pixel-by-pixel basis. The fuel was a mixture of acetaldehyde diluted 1/1 by air (on a mass basis) to eliminate soot. It emanated from a piloted burner ($d = 3.9$ mm) with a velocity corresponding to $Re = 18,000$. The flame was enshrouded in a low velocity filtered air coflow in order to keep the measurement area clear of particles, which would interfere with the Rayleigh images.

A conserved scalar, β , based on the fuel mass fraction (Y_f) and the enthalpy ($H = c_p T/Q$) served as the basis for determining the mixture fraction:

$$\beta = Y_f + c_p T/Q$$

$$\xi \equiv \frac{\beta - \beta_2}{\beta_1 - \beta_2} = \frac{Y_f + c_p/Q(T-T_2)}{Y_{f,1} + c_p/Q(T_1-T_2)}$$

where the subscripts denote the fuel (1) and oxidizer (2) streams. Y_f is obtained from the fluorescence images and T is related to the Rayleigh images. The mixture fraction can be expressed in terms of the fluorescence signal, F , and the Rayleigh signal, R_a , as follows:

$$\xi = \frac{C_1 a_T F}{W R_a F_{cal}} + C_2 (a_T/R_a - T_{ambient})$$

where C_1 , C_2 , and F_{cal} are calibration constants, a_T is proportional to the local Rayleigh cross section, and W is the local mixture molecular weight. Since a_T and W are both functions of ξ , an iterative process was used on a pixel by pixel basis to derive the

temperature and mixture fraction. The procedure converges quickly, usually within fewer than four iterations.

Preliminary data on the calculated temperature and mixture fraction obtained from the instantaneous Rayleigh/fluorescence images show several interesting features. Evident in the temperature mappings are the high temperature zones on the outer edge of the flame. As expected, the mixture fraction peaks in the unburned regions near the center of the jet. Closer investigation of the mixture fraction images reveals a slight decrease in signal around the unburned gas on the fuel-rich side of the flame front. This loss of signal could be due to a variety of factors. As the acetaldehyde approaches the flame front, the rapid increase in temperature may cause the acetaldehyde to pyrolyze, thereby eliminating the fluorescence. Assuming a one step reaction from fuel to products may also be the problem: when the parent fuel is converted to intermediates, the fluorescence is lost. Thus, the premature loss of signal may be responsible for the dip in the mixture fraction. Preliminary studies in our laboratory in a laminar diffusion flame show a pronounced dip in the mixture fraction at the flame front. With the increased residence time in the laminar flame, pyrolysis effects are expected to predominate. In the turbulent flame studied, however, residence times are short so pyrolysis effects may be only one part of the problem witnessed.

Photodissociation is another possible source of the loss of signal near the flame front. Although 320 nm is above the dissociation wavelength at room temperature, the high temperature in the flame may alter this threshold. We have found that 320 nm excitation has significantly less dissociation in the flame than 280 nm excitation, but further experiments are needed to ascertain the optimum excitation wavelength.

The results of this preliminary study show significant promise in terms of obtaining useful measurements of the mixture fraction. The strength of the signals from the Rayleigh scattering and the fuel fluorescence allows computations of the mixture fraction high in spatial resolution and low in noise, thus making determination of the scalar dissipation

feasible. The main question raised by our preliminary experiments relates to the adequacy of the acetaldehyde fluorescence as a means of determining the fuel concentration. As stated above, there are indications that near the flame front, there is dropout of the fluorescence signal. It is currently not clear whether this is due to pyrolysis, photodissociation, or the single-step chemistry assumption. One means of investigating this will be to do simultaneous Raman and fluorescence measurements in the flame. The Raman signal from C-H stretch may be less sensitive to pyrolysis and, if done with a different laser, is independent of any photodissociation caused by the fluorescence laser. The results of these experiments should clarify the source of the drop in the fluorescence signal and may make it possible to eliminate it by changing the fluorescence excitation wavelength or to compensate for it in the mixture fraction calculation.

If the acetaldehyde fluorescence technique proves to be unacceptable near the flame front, there are other possibilities for measuring the fuel concentration. For example, other fuel tags such as acetone may prove better than acetaldehyde. The temperature dependence of the acetone fluorescence is known, making it easier to correct the signal in high temperature regions. Alternatively, monitoring the C-H Raman signal from a hydrocarbon fuel such as methane will yield the fuel concentration field directly. Although the process is weaker than the fluorescence approach, it has the advantages that there is no quenching or dissociation to cope with in the data analysis. With the new technology in intensified CCD detectors available, coupled with the high-energy dye laser already in use at our facility, it may be feasible to obtain high quality mixture fraction data from Rayleigh/Raman imaging. In any case, we will investigate which fuel/fuel tag and excitation-wavelength scheme is optimum.

Our work on conserve scalar imaging in turbulent nonpremixed flames is still in its early phases and most of the results are unpublished, however, the first of our preliminary results on conserved scalar imaging have been submitted for publication and new results

will be presented at invited lectures at CLEO '93 and at the upcoming Gordon Conference on the Physics and Chemistry of Laser Diagnostics in Combustion.

(2) Other Research Efforts

In addition to initiating new conserved scalar imaging experiments, we have continued development in the application of the diagnostic technique we have introduced previously. The milestones related to these other diagnostic techniques within the last year include the following:

- (a) We have conducted an initial investigation into the use of computer aided tomography for three-dimensional measurements in a flame. In these experiments five cameras were used to measure the flame luminosity of a methane diffusion flame. Tomographic reconstruction techniques were used to obtain the three-dimensional light intensity distribution of the luminous zone. This work was done in collaboration with Dr. Dean Verhoeven at Institut Francais du Petrole.
- (b) Presentation of our previous scalar imaging work at the Twenty-Fourth (Int'l) Symposium on Combustion in Sydney, Australia.
- (c) Acceptance of our previously submitted paper on differential diffusion.
- (d) Simultaneous PIV and scalar measurements were made in premixed flames. Our effort during the past year has specifically involved some preliminary measurements of joint PIV and CH fluorescence measurements in flames. In addition, progress has been made on directional ambiguity resolution in PIV studies. Future plans involve development of techniques to measure the all three components of the velocity field.

PUBLICATIONS RESULTING FROM THE RESEARCH

I. Nonlinear Spectroscopy of Multicomponent Droplets

1. A. Serpengüzel, G. Chen, R. K. Chang, and W.-F. Hsieh, "Heuristic Model for the Growth and Coupling of Nonlinear Processes in Droplets," *JOSA B* **9**, 871 (1992).
2. R. K. Chang and Ali Serpengüzel, "Characteristics and Applications of Stimulated Raman Scattering in Microdroplets," in Proceedings of the XIIIth International Conference on Raman Spectroscopy, 1992, Würzburg, Germany, W. Kiefer, M. Cardona, G. Schaack, F. W. Schneider, and H. W. Schrötter, eds. (John Wiley and Sons, Chichester, 1992), p.33.
3. David H. Leach, Richard K. Chang, William P. Acker, and Steven C. Hill, "Third Order Sum Frequency Generation in Droplets: Experimental Results," *JOSA B* **10**, 34 (1993).
4. Steven C. Hill, David H. Leach, and Richard K. Chang, "Third Order Sum Frequency Generation in Droplets: Model with Numerical Results for Third Harmonic Generation," *JOSA B* **10**, 16 (1993).
5. Ali Serpengüzel, Richard K. Chang, William P. Acker, and Rodney L. Sung, "Laser Diagnostic Techniques for Characterizing Droplet Size, Composition, and Differential Evaporation in Fuel Sprays," in Proceedings of the XXIV FISITA Congress, 1992, London, England, paper number 925030, (Mechanical Engineering Publications Limited, London, 1992) p. 107.
6. Alfred S. Kwok and Richard K. Chang, "Fluorescence Seeding of Weaker-Gain Raman Modes in Microdroplets: Enhancement of Stimulated Raman Scattering," *Opt. Lett.* **17**, 1262 (1992).
7. J. Christian Swindal, David H. Leach, Richard K. Chang, and Kenneth Young, "Precession of Morphology-Dependent Resonances in Non-Spherical Droplets," *Opt. Lett.* **18**, 191 (1993).
8. Gang Chen, J. Christian Swindal, and Richard K. Chang, "Frequency Splitting and Precession of Cavity Modes of a Droplet Deformed by Inertial Forces," in Proceedings of Shanghai International Symposium on Quantum Optics, Vol. 1726 (SPIE, Bellingham, Washington, 1992) p. 292.
9. Gang Chen, Ali Serpengüzel, Richard K. Chang, and William P. Acker, "Relative Evaporation of Droplets in a Segmented Stream Determined by Cavity Droplet Fluorescence Peak Shifts," in Proceeding of the SPIE Conference on Laser Applications in Combustion and Combustion Diagnostics, Los Angeles, CA, January 18-23, 1993 (to be published).
10. Alfred S. Kwok and Richard K. Chang, "Suppression of Lasing by Stimulated Raman Scattering in Microdroplets," Extended Abstract of Joint Meeting of CLEO and QELS, Baltimore, MD, May 3-7, 1993 (to be published).

II. Two- and Three-Dimensional Measurements in Flames

11. S. H. Stårner, R. W. Bilger, K. M. Lyons, D. F. Marran and M. B. Long, "Planar Mixture Fraction Measurements in Turbulent Diffusion Flames by a Joint Rayleigh and Fuel LIF Method," submitted to the Fourteenth International Colloquium on the Dynamics of Explosions and Reactive Systems, 1993.
12. S. H. Stårner, R. W. Bilger, R. W. Dibble, R. S. Barlow, D. C. Fourquette and M. B. Long, "Joint Planar CH and OH LIF Imaging in Piloted Turbulent Jet Diffusion Flames Near Extinction," in Proceedings of the Twenty-Fourth Symposium (International) on Combustion, Sydney, Australia, July 5-10, 1992, (in press).
13. M. B. Long, S. H. Stårner, and R. W. Bilger, "Differential Diffusion in Jets Using Joint PLIF and Mie Imaging," Comb. Sci. Tech. (1992) (in press).
14. K. M. Lyons, J. H. Frank and M. B. Long, "Simultaneous Velocity and Concentration Measurement in a Gas Phase Jet Using Digital PIV and LIF Imaging," to be submitted to Applied Optics.

SCIENTIFIC COLLABORATORS

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

I. Nonlinear Spectroscopy of Multicomponent Droplets

Outside Scientists: William P. Acker (Texaco)
Rodney L. Sung (Texaco)
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Steven C. Hill (U.S. Army Atm. Sciences Lab.)
Kenneth Young (Chinese Univ. of Hong Kong)
David H. Leach (Max-Planck-Institut., Stuttgart)
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Post Doctoral Scientist: Ali Serpengüzel

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II. Two- and Three-Dimensional Measurements in Flames

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LECTURES PRESENTED ABOUT THE RESEARCH

Richard K. Chang: (Talks not delivered by Richard Chang are designated by *).

* "Frequency Splitting and Precession of Cavity Modes of a Droplet Deformed by Inertial Forces," Shanghai International Symposium of Quantum Optics, Shanghai, China, March 30-April 2, 1992. (Delivered by Gang Chen).

"Nonlinear Optics in Single Droplets," Physics Department Seminar, Harvard University, Cambridge, MA, April 3, 1992.

"Basic Studies and Applications of Nonlinear Emission from Microdroplets," Physics Department, The Chinese University of Hong Kong, Shatin, HK, April 13-15, 1992. (Series of three lectures).

* "Laser Diagnostic Techniques for Characterizing Droplet Size, Composition, and Differential Evaporation in Fuel Sprays," FISITA Congress, London, England, June 7-11, 1992. (Delivered by William P. Acker).

"Nonlinear Spectroscopy of Multicomponent Droplets," AFOSR Propulsion Contractors Meeting, La Jolla, CA, June 15-19, 1992.

"Stimulated Emission from Microdroplets" The Second Workshop on Quantum Optics, Weizmann Institute of Science, Rehovot, Israel, June 22-26, 1992. (Invited talk).

* "Precession of Morphology-Dependent Resonances in Non-Spherical Droplets," (delivered by J. Christian Swindal); and "Fluorescence Seeding of Stimulated Raman Scattering in Droplets," (delivered by Alfred S. Kwok), Scientific Conference on Obscuration and Aerosol Research, U.S. Army Chemical Research, Development and Engineering Center, Aberdeen Proving Ground, MD, June 22-26, 1992.

"Nonlinear Optical Effects in Micrometer Size Particles, I & II," 5th International Topsoe Summer School on Nonlinear Optics, Aalborg, Denmark, August 3-8, 1992. (Two invited talks).

* "Fluorescence Seeding of Stimulated Raman Scattering in Microdroplets," Topical Meeting on the Nonlinear Optics: Materials, Fundamentals, and Applications, Maui, Hawaii, August 17-21, 1992. (Delivered by Alfred S. Kwok).

"Determination of Chemical and Physical Properties of Droplets from Nonlinear Spectroscopy," Edwards Air Force Base, CA, August 21, 1992.

"Characteristics and Applications of Stimulated Raman Scattering in Microdroplets", XIIIth International Conference on Raman Spectroscopy, Würzburg, Germany, August 31- September 4, 1992. (Plenary lecture).

"Nonlinear Optics inside a Microdroplet," Atomic Physics Seminar, State University of New York, Stony Brook, NY, October 14, 1992.

"Chemical Speciation of a Droplet with Nonlinear Optical Effects," Physics Seminar, RPI, Rensselaer, NY, October 14, 1992.

* "Third Order Sum Frequency Generation in Droplets," Annual Meeting of the OSA, Albuquerque, NM, September 20-25, 1992. (Delivered by Steven C. Hill).

* "Status Report of Nonlinear Diagnostics of Droplets and Sprays," UTRC, East Hartford, CT, November 11, 1992. (15 minutes talks delivered by 5 students and 1 post doctoral scientist).

"Evaporation of Droplets as Detected by Stimulated Light Scattering," Air Force Aero-Propulsion Laboratory, Wright Patterson Air Force Base, Dayton OH, December 2, 1992.

"Determination of Chemical and Physical Properties of Droplets from Nonlinear Spectroscopy," NASA Langley, Cleveland, OH, December 3, 1993.

* "Relative Evaporation Rates of Droplets in a Segmented Stream Determined by Droplet Cavity Fluorescence Peak Shifts," SPIE Conference on Laser Applications in Combustion and Combustion Diagnostics, Los Angeles, CA, January 18-23, 1993. (Delivered by Ali Serpengüzel).

Marshall B. Long

"Conserved Scalar Measurements in a Turbulent Nonpremixed Flame," presented at The 19th Sandia Turbulence in Aerothermochemistry Research (STAR) Meeting, held at the Lawrence Berkeley Laboratory, Berkeley, California, April 1992.

"Imaging Measurements in a Combustion Bomb," presented at Texaco, Beacon, New York, January, 1993.

"Joint Planar CH and OH LIF in Piloted Turbulent Jet Diffusion Flames Near Extinction," Twenty-Fourth Symposium (International) on Combustion, Sydney, Australia, July, 1992.

INTERACTIONS WITH OTHER LABORATORIES

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CRDEC, Aberdeen Proving Ground, MD
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Colorado State University, Fort Collins, CO
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DEGREES AWARDED

Ali Serpengüzel, Ph. D., May, 1992.

Kevin M. Lyons, M. Phil., May, 1992.

PATENTS

None