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19. Abstract (Continued)

In the experiments on two- and three-dimensional measurements in turbulent flames, progress is reported in the following areas: (1) The temporal development of three-dimensional flow structures has been recorded by performing a series of planar measurements in an acoustically forced jet flow. (2) The evolution of the flame front in a turbulent hydrogen-air premixed flame was recorded at a 48 kHz rate, which allowed measurement of the convection and burning velocities. (3) An experiment is described on the measurement of all three components of the scalar gradient at points within a plane intersecting a flow. The data from this experiment have been used by other researchers to advance a new theory of turbulent non-premixed flames and have led to a collaboration on laser diagnostics development.

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Annual 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
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NONLINEAR SPECTROSCOPY OF MULTICOMPONENT DROPLETS

RESEARCH OBJECTIVES

Following is a brief description of the three principal research objectives related to nonlinear spectroscopy of liquid droplets:

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 droplets.

RESEARCH STATUS

Our results for the first year of research on nonlinear spectroscopy of droplets can be summarized as follows:

1. Laser-Induced Shape Deformation by Electrostriction

We began with the last research objective listed above. 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 at the internal focal spot just within the droplet shadow face. The laser-induced electrostrictive force pushes against the surface tension force of the droplet which bulges at the droplet shadow face. The maximum amount of shape distortion is proportional to the electrostrictive impulse, which is a linear function of $[\nabla I \times (\Delta\tau)]$, where $\Delta\tau$ is the laser pulse duration and $I \times (\Delta\tau)$ is the laser energy. At high input laser energies, the bulge acquires a cylindrical shape and smaller droplets are noted to break away from this liquid column before the bulge contracts. 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 dynamic surface tension of the liquid, and the dampening rate is 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. Since the SRS intensity threshold is dependent on the MDR's that provide the necessary optical feedback, any shape distortions will result in lowering the Q factor associated with the MDR's. Although the droplet shape deformation is too small to observe visually when the laser and SRS pulses are on, the lowering of the Q factor can still be significant. Consequently, the higher the input laser intensity is, the less SRS is generated once laser-induced shape distortion is large enough to

degrade the Q factor of the MDR, which is within the Raman gain profile. [Our results have been reported in Optics Letters (publication #1).]

2. 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. Laser-induced breakdown 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. [Our results have been reported in Optics Letters (publication #2).]

3. Excitation of SRS with Single-Mode and Multimode Q-Switched Lasers

We initiated the experimental study of the SRS statistics from single droplets in a flowing stream (our first research objective). The SRS intensity threshold was investigated with a single-mode and a multimode Q-switched Nd:YAG laser. The second harmonic beam of a single- or multimode Q-switched Nd:YAG laser is tightly focused at the center of the droplet illuminated face in order to avoid exciting any MDR's, regardless of the laser wavelength or droplet radius.

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) with single-mode excitation, where the laser linewidth is less than the spontaneous Brillouin linewidth, the intensity threshold for stimulated Brillouin scattering (SBS) from droplets is lower than that for SRS; (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 SRS pulse occurs when the n th SRS pulse reaches a maximum.

We have tentatively concluded that, for single-mode laser excitation of droplets, the SRS is pumped by the internal SRS and not by the internal radiation of the laser. This conclusion has a profound effect on the SRS fluctuations which affect the accuracy of our determination of the concentration of multicomponent liquid droplets in a spray. [Our findings will be submitted to the J. Opt. Soc. Am. B for publication.]

4. Fluorescence Imaging of Droplets with a CCD Detector

As a prelude to using CARS spectroscopy to determine the concentration distribution from different regions within the droplet rim with a CCD two-dimensional detector (research objective 2), we initiated studies on fluorescence imaging from different regions within the droplet rim. The fluorescence signals are known to be several orders of magnitude larger than the CARS signals.

Using the *excimer and monomer fluorescence technique* which Prof. Lynn Melton has developed for bulk liquids, we initiated a program to determine the internal temperature distribution of the droplet. A CO_2 laser pulse (propagating in the z direction) irradiates a droplet and heats the ethanol droplet at its shadow face where the CO_2 laser beam is focused. A N_2 laser pulse (propagating in the $-z$ direction) irradiates the same droplet and causes the droplet to emit its excimer fluorescence (green when pyrene is used) and monomer fluorescence (blue when pyrene is used). The excimer and monomer fluorescence images are focused on the upper and lower portions of the CCD detector, respectively. The monomer and excimer intensities are then divided pixel-by-pixel, $I_{\text{monomer}}(x,y)/I_{\text{excimer}}(x,y)$, which is proportionally related to the temperature distribution within the droplet $T^*(x,y)$. By delaying the N_2 laser pulse relative to the CO_2 laser pulse, the time development of $T^*(x,y)$ can, in principle, be determined. The spatial distribution of the measured fluorescence intensity ratio was compared with the calculated internal intensity distribution for an ethanol droplet of radius a and complex index of refraction at the CO_2 laser wavelength. We also calculated the fluorescence "escape" efficiency for a dipole emitting in a plane containing the north

and south poles of the droplet.

The results were quite irreproducible because of the time varying spatial distribution of the CO₂ laser beam during each pulse. Consequently, different portions of the droplet were nonuniformly heated by the CO₂ laser beam during each pulse. This experimental setback could be overcome by using a CO₂ laser with a more uniform spatial distribution. A more serious problem is associated with droplet shape distortion caused by the lowering of the surface tension as the droplet illuminated face is heated by the CO₂ laser beam. Shape distortions cause the fluorescence escape efficiency to change drastically. Unfortunately, chromatic aberrations, which lead to a false $T^*(x,y)$, cannot be eliminated by dividing the fluorescence intensity distributions in the green (from the excimer) and in the blue (from the monomer). [Prof. Melton and our group will continue to try to overcome some of the difficulties related to the technique.]

In the process of attempting to map the internal temperature distribution within a droplet via the excimer-monomer fluorescence technique, we realized that the normal fluorescence (monomer) imaging technique can be used to determine the shape of the droplet in the liquid phase. Demarcation of the liquid and vapor phases is evident in the fluorescence intensity distribution, which is intense for the liquid phase and weak for the vapor phase, consistent with the dye molecular density in the liquid and vapor phases. The intensity contrast between the two phases was particularly distinct when the laser-induced vaporization left the dye molecules in the remaining droplet and only ethanol was partially vaporized. By detecting the fluorescence images at different time delays after CO₂ laser beam irradiation, the shapes of the remaining droplet are imaged without much background fluorescence from the vapor. Because of the dark background in regions outside the remaining droplet, small droplets are detected to break away from the parent droplet.

The fluorescence imaging technique is much superior to the conventional shadowgraph technique used by us and other groups to image the shape of the droplet at various time delays after the CO₂ laser beam irradiation. The contrast between the liquid and vapor phases is poor with the shadowgraph approach while it is excellent with the fluorescence approach. [We are in the process of plotting the intensity distribution (in grey scale) for submission to Optics Letters.]

PUBLICATIONS

1. J.-Z. Zhang and R.K. Chang, "Shape Distortion of a Single Water Droplet by Laser-Induced Electrostriction," Opt. Lett. 13, 916 (1988).
2. 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).

PROFESSIONAL PERSONNEL

Jian-Zhi Zhang, Graduate Student

Carol F. Wood, Research Associate

DEGREES AWARDED

None

PATENTS

None

COUPLING ACTIVITIES

Some of the temperature mapping and SRS results were presented at the following meetings and workshops:

Workshop on Mass, Momentum, and Energy Exchange in Combusting Spray:
Droplet Studies
Sandia National Laboratories, Livermore, California
March 28-29, 1988

JANNAF Meeting on Kinetics and Related Aspects of Propellant Combustion Chemistry
Applied Physics Laboratory, John Hopkins University, Laurel, Maryland
May 2-3, 1988

17th DOE/Diesel Cooperative Working Group Meeting
Southwest Research Institute, San Antonio, Texas
May 19-20, 1988

AFOSR Contractors Meeting on Rocket Propulsion
Monrovia, California
June 14-15, 1988

1988 CRDEC Scientific Conference on Obscuration and Aerosol Research
CRDEC, Edgewood Arsenal, Maryland
June 22-23, 1988

4th Annual Conference on HAN-Based Liquid Propellant Structure and Properties
Ballistic Research Laboratory, Aberdeen Proving Ground, Maryland
August 30-31, 1988

XIII International Conference on Coherent and Nonlinear Optics
Minsk, USSR
September 6-9, 1988

18th DOE/Diesel Cooperative Working Group Meeting
Penn State University, University Park, Pennsylvania
November 17-18, 1988

TWO- AND THREE-DIMENSIONAL MEASUREMENTS IN FLAMES

RESEARCH OBJECTIVES

The overall objectives of the work on the development of laser diagnostic techniques for two- and three-dimensional measurements in flames are as follows:

1. To refine and extend three-dimensional measurement techniques. The ability to obtain three-dimensional measurements in turbulent flows has been demonstrated. More work is required, however, to solve problems specific to these measurements and to improve their accuracy. Aspects to be addressed include increasing signal/noise, minimizing geometric distortions, and improving the spatial registration of the set of two-dimensional images that form the three-dimensional measurement. If the problems can be solved, the ability to characterize fully the topology of structures in the flow will be realized. Quantities such as surface/volume ratios, gradients, curvatures, and fractal dimensions can all be determined from these three-dimensional data sets.
2. To measure 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, rates of 5 - 50 kHz are required to follow the evolution of structures.
3. To measure 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. The image pair obtained contains the information required to determine all three components of the gradient vector at each point within the plane. From a large ensemble of instantaneous measurements, it is possible to determine the joint pdf of the scalar and its gradient. This information can be compared directly with current combustion models.

RESEARCH STATUS

During the first year, progress in the development and application of laser diagnostics for two- and three-dimensional measurements includes the following:

1. Measurement of the Time Development of 3-D Structures in Forced Flows

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. [Some of our work on three-dimensional measurements was reported at the 22nd Symposium (International) on Combustion (publication #1).]

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 has been submitted to the journal *Computer* for inclusion in a special issue related to visualization in scientific computing (publication #2).

2. Temporal Evolution of a Premixed Hydrogen-Air Flame

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 written and will be submitted to Combustion Science and Technology (publication #3).]

3. 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 submitted for publication (publication #4).

4. Simultaneous CH and CH₄ Mapping

Another collaborative experiment was done with researchers at Sandia National Laboratories in Livermore, California. In this work, a technique has been 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 this experiment have been reported in Applied Optics (publication #5).

PUBLICATIONS

1. M.B. Long and B. Yip, "Measurement of Three-Dimensional Concentrations in Turbulent Jets and Flames," in *Proceedings of the Twenty-Second Symposium (International) on Combustion*, The Combustion Institute, in press.
2. M.B. Long, K. Lyons, and J.K. Lam, "Acquisition and Representation of Two- and Three-Dimensional Data from Turbulent Flows and Flames," submitted to *Computer*.
3. M. Winter and M.B. Long, "Two-Dimensional Measurements of the Time Development of a Turbulent Premixed Flame," to be submitted to *Combust. Sci. Tech.*
4. R.W. Bilger, B. Yip, M.B. Long, and A.R. Masri, "An Atlas of QEDR Flame Structures," submitted to *Combust. Sci. Tech.*
5. 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).

PROFESSIONAL PERSONNEL

Michael Winter, Graduate Student

Brandon Yip, Graduate Student and Research Associate

DEGREES AWARDED

Brandon Yip, "Three-Dimensional Laser Diagnostics in Gas Flows," Ph.D. (1988).

Michael Winter, "Two-Dimensional Measurements of the Time Development of Gas-Phase Flows," Ph.D. (1988).

PATENTS

None

COUPLING ACTIVITIES

Presentations about the work on two- and three-dimensional measurements in flames were made at the following meetings and workshops:

AFOSR Contractors Meeting on Rocket Propulsion
Monrovia, California
June 14-15, 1988.

Twenty-Second Symposium (International) on Combustion
University of Washington, Seattle, Washington
August 14-19, 1988.

Department of Mechanical Engineering Seminar Series
Cornell University, Ithaca, New York
September, 1988.

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.

In addition, our collaboration with researchers at Sandia National Laboratories in Livermore, California has continued.