INVESTIGATION TO EXTEND THE APPLICABILITY OF
LASER RAMAN SCATTERING DIAGNOSTIC
TECHNIQUES TO PRACTICAL COMBUSTION SYSTEMS

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ALAN C. ECKBRETH
UNITED TECHNOLOGIES RESEARCH CENTER
EAST HARTFORD, CONNECTICUT 06108

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PROJECT SQUID HEADQUARTERS
CHAFFEE HALL
PURDUE UNIVERSITY
WEST LAFAYETTE, INDIANA

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United Technologies Research Center
East Hartford, Connecticut 06108
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Alan C. Eckbreth, Principal Investigator

Introduction

With the development of high power, visible laser sources, Raman scattering techniques have received considerable attention for combustion diagnostics in the past several years. Laser Raman approaches have been advanced to the point where they are now being routinely employed in a variety of fundamental flame investigations. However, widespread application of laser Raman techniques to practical combustion devices such as gas turbines, hydrocarbon combustors, furnaces, etc., is yet to be achieved. From an instrumentation standpoint, practical devices contain flames which differ markedly from those typically employed in fundamental laboratory studies. Environments of practical interest contain flames which are generally turbulent, high luminous and particulate laden. The high luminosity levels preclude the use of cw laser sources for Raman work in these environments. Pulsed laser sources are required to produce peak Raman powers comparable to or preferably in excess of the background luminous power. In some instances, low S/N ratios are encountered even with pulsed lasers. In situations where the background luminosity is tolerable, the pulsed laser-particulate interaction can lead to a
variety of laser-particulate interaction "noise" effects which can adversely affect the successful application of laser Raman techniques. In laser Raman thermometry experiments at UTRC, successful Raman measurements in a hydrocarbon-fueled model combustor were, in one case, precluded by laser modulated particulate incandescence. Particles (e.g., soot) in the measurement volume, which at high stream temperatures are already incandescent and contribute significantly to the total luminous emission, absorb the incident Raman-inducing laser radiation, heat to temperatures far above ambient, and emit greatly increased amounts of gray/blackbody radiation which can exceed the sought-for Raman signal levels. Under Project SQUID sponsorship, this problem and potential solutions have been experimentally investigated under controlled laboratory conditions. Details of these studies will soon be available in a Project SQUID technical report (Ref. 1).

Discussion

Detailed analyses (Ref. 2) of laser irradiated particle heating have been made in an effort to obtain a better physical understanding of this phenomena and to seek solutions to laser induced or modulated incandescence "noise" effects. In general, these analyses display the inadequacy of the various heat transfer processes from preventing the surface temperature from rising appreciably above the initial level, particularly for larger particle sizes, i.e., \( \geq 1 \mu \), and higher focal flux values, i.e., above \( 10^5 \text{ W/cm}^2 \). Due to this inadequacy, the particle surface temperature tends to follow the laser pulse envelope exhibiting little phase behavior which could be exploited for noise suppression. Despite the high
surface temperature produced, the analysis predicts a relatively weak dependence of surface temperature on focal flux. Thus, even though the surface temperature and the incandescence rises with increasing focal flux, the incandescent noise does not increase as rapidly as the Raman signal, and S/N improvements are predicted with increasing levels of focal flux.

Experimental Laser Irradiated Particle Noise Studies This aspect of the investigation addressed the validity of foregoing analysis. Initially laser induced particulate noise was studied by irradiating singly suspended 5 μm diameter particles. Early results, including the presence of laser induced C₂ Swan emission in addition to incandescence, were summarized previously (Ref. 3). One important point worth reemphasizing concerns the distinction between alumina and carbon (soot) particulates which has been experimentally verified. Unlike soot particles, Al₂O₃ possesses an extremely low absorption cross section, which varies from four to five orders of magnitude less than its geometric cross section (Ref. 4) depending on particle size and wavelength. Consequently, very little incident laser energy is adsorbed by an alumina particle resulting in very little heating and, hence, very little laser induced radiative noise. Hence, Al₂O₃ would be an excellent seed for LDV work permitting simultaneous LDV and laser Raman studies.

Particle size measurements in the UTRC test combustor (Ref. 5) using a Mie scattering technique indicated an average particle size of 400 μm (0.04 μm). Consequently, subsequent testing focussed on laser induced particulate noise produced by laminar propane diffusion flames whose soot sizes were known to be in this range. Laser induced particulate radiation was monitored as a function of energy
and focal flux employing a two channel Raman spectrometer (Ref. 2). For these studies, the spectrometer functioned as a two color pyrometer (5230, 6864 Å) and was calibrated against a tungsten filament as known temperature. Laser heated particulate temperatures, determined in the above manner, as a function of focal flux level exhibited fairly good agreement with the analytical model (Ref. 2). The particulate temperatures varied from approximately 3500°K to 4500°K over a flux range from $10^5$ to $10^8$ W/cm$^2$. Quite importantly, the absolute noise level after increasing initially with laser energy, leveled off and became nearly independent of energy for laser energies in excess of about 60 mJ. This results essentially from the saturable behavior of the laser heated particle surface temperature with increasing focal flux. The laser induced particulate noise also decreased with decreasing focal length due to the decrease in the number of particles being irradiated. From this data, a signal to noise improvement of several hundred was extrapolated for flux increases from $2(10^5)$ W/cm$^2$ to $10^9$ W/cm$^2$. Operation beyond this flux level may be problematical due to gas breakdown. These results clearly indicate the value of operating at the highest possible focal flux level in particulate laden environments. Operation at these flux levels can lead to other problems, however, such as optical damage in and fluorescence from window ports, both of which have been experimentally encountered.

**Signal Averaging Considerations** In Ref. 6, the consequences of time averaging Raman data generated by a cw laser from a turbulent medium were examined. In an analogous fashion, an analysis of ensemble averaging pulsed Raman data in a fluctuating medium was performed. The analysis can handle such effects as laser
energy variations and shot noise fluctuations and treats among other items, the case where "noise" is sampled and subtracted from a "signal" channel. The analysis demonstrated that when Raman data are averaged, background can be subtracted on average and shot noise terms average to zero. However, temperature inferred from the ratio of separately average band intensities will not be the average temperature but will depart from the true average depending on the magnitude and correlation of the fluctuations. Furthermore, in measurement situations where the band intensity ratio at any instant cannot be determined accurately (due to shot noise effects, including those arising during subtraction), the average of these separate erroneous temperature measurements will not yield the true average temperature.

**Seeded Flame Temperature Measurements** A CH₄ diffusion flame sustained in the center of a 7.6 cm dia. premixed flat flame burner was employed to introduce controlled amounts of laser induced particulate noise into a Raman temperature measurement experiment. A four channel Raman spectrometer consisting of two signal and two noise channels, the latter located in spectral regions just adjacent to the Raman bands, was used to demonstrate noise sampling and subtraction. Due to shot noise fluctuations, good subtraction is not obtainable with each pulse, but only on average over a large number of pulses depending on the photon levels involved. By adjusting the noise channel photomultipliers in ten volt increments good subtraction was obtained on average with the dye laser so tuned that all four channels "saw" only noise. By appropriate tuning of the laser, the Raman bands were placed within the signal bandpasses and observed with the noise subtracted
out. These signals were compared with Raman data with the seed flame extinguished and agreed quite well since the introduction of the seed flame constituted only a small perturbation on the burner temperature profile. Thus noise sampling and subtraction is a viable approach for Raman measurements in noisy media subject to potential errors arising from signal averaging in unsteady environments.

Conclusions and Recommendations. It is difficult to be categorical in regard to the utility of spontaneous Raman scattering for practical combustion diagnostics since conditions can vary widely from device to device and within a given device, depending on location and operating parameters. Based upon our studies, primary zone diagnostics in a diffusion flame apparatus operating on hydrocarbon fuels appears highly doubtful based upon both background luminosity considerations and laser induced particulate noise effects. Secondary and exhaust region probing may be possible (Ref. 7) since luminosity levels tend to be lower, but particulate effects could be problematical if the particles are in sufficient quantity or size. High focal flux levels appear optimal for enhancing Raman S/N in regard to laser induced particulate noise but may lead to problems with window breakdown and fluorescence. The latter may be sufficiently strong to preclude the use of preferred background luminosity suppression geometries. Noise sampling and subtraction is feasible as has been demonstrated. However, shot noise effects dictate that Raman data be separately averaged to permit accurate determination of average signal levels. Ensemble averaging, however, leads to band intensity ratios which, in fluctuating environments, depend not only on average temperature,
but on average density and on the magnitude and correlations of fluctuations in
density and temperature. These effects may result in large measurement errors
rendering the extracted Raman data of little utility. Based on this perception
of the situation, we believe spontaneous Raman scattering to be of limited utility
for many practical applications. However, due to its relative simplicity and
high level of present day understanding it certainly merits first consideration
in environments where the aforementioned noise effects are not overwhelming.

For most practical applications, the feasibility of stronger Raman processes
such as CARS (coherent anti-Stokes Raman scattering) or near-resonant Raman
scattering needs to be investigated. In particular CARS has shown great potential
to date but is not without problems due to its sophistication. It is not completely
clear at this point what the impact of practical device characteristics such as
high turbulence and particulate levels will be on CARS generation. In particular
since nonresonant susceptibility contributions are often problematical in CARS
experiments, potential particulate (soot) noise effects should be examined in a
controlled manner.

References

1. Eckbreth, A. C.: "Investigation to Extend the Applicability of Laser Raman
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2. Eckbreth, A. C.: "Laser Raman Thermometry Experiments in Simulated Combustor
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Laser Raman scattering diagnostic techniques may be precluded from application to practical combustion devices due to a variety of laser-particulate (soot) interaction "noise" effects. Under the subject contract the laser particulate interaction has been studied in some detail. Laser induced particulate temperatures have been measured as a function of laser flux and are found to agree fairly well with an analytical model. The scaling of the absolute noise...
20. has been studied and the noise has been found to saturate with increasing laser energy and decrease with decreasing focal length lenses. The effect of signal averaging pulsed Raman data obtained in an unsteady medium has been analyzed. Averaged Raman data is shown to depend not only on average temperature but on the magnitudes and correlations of medium fluctuations in density and temperature. Noise sampling and subtraction have been shown to be feasible and a temperature measurement demonstration was performed on a soot seeded laboratory burner using the above approach.