1995

NASA/ASEE SUMMER FACULTY FELLOWSHIP PROGRAM

MARSHALL SPACE FLIGHT CENTER
THE UNIVERSITY OF ALABAMA IN HUNTSVILLE

Thermodynamic Measurements in a High Pressure Hydrogen-Oxygen Flame Using Raman Scattering from a Broadband Excimer Laser

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Introduction

Raman scattering is an inelastic molecular scattering process in which incident radiation is reemitted at a fixed change in frequency.[1] Raman spectroscopy can be used to measure the number density and temperature of the irradiated species.[2] The strength of the Raman signal is inversely proportional to the wavelength raised to the fourth power. Consequently, high signal to noise ratios are obtained by using ultraviolet (UV) excitation sources. Using UV sources for Raman Spectroscopy in flames is complicated by the fact that some of the primary constituents in hydrogen-oxygen combustion absorb and reemit light in the UV and these fluorescence processes interfere with the Raman signals.[3] This problem has been handled in atmospheric pressure flames in some instances by using a narrowband tunable excimer laser as a source. This allows for detuning from absorption transitions and the elimination of interfering fluorescence signals at the Raman wavelengths.[4] This approach works well in the atmospheric pressure flame; however, it has two important disadvantages. First, injection-locked narrowband tunable excimer lasers are very expensive. More importantly, however, is the fact that at the high pressures characteristic of rocket engine combustion chambers, the absorption transitions are broadened making it difficult to tune to a spectral location at which substantial absorption would not occur.

The approach taken in this work is to separate the Raman signal from the fluorescence background by taking advantage of the fact that Raman signal has nonisotropic polarization characteristics while the fluorescence signals are unpolarized. Specifically, for scattering at right angles to the excitation beam path, the Raman signal is completely polarized. The Raman signal is separated from the fluorescence background by collecting both horizontally and vertically polarized signals separately. One of the polarizations has both the Raman signal and the fluorescence background while the other has only the fluorescence signal. The Raman scatter is the difference between the signals. By choosing an appropriate optical setup, both signals can be obtained simultaneously with the same monochromator; hence, time resolved measurements are possible using this approach.

An alternative approach for using the polarization characteristics of the Raman scattering process to separate the Raman signal from the fluorescence background is to take advantage of the directional dependence of the scattering on the laser polarization. For a laser polarization in the plane formed by the laser beam and the collection path, no Raman scatter will be detected for a collection path which is perpendicular to the incident radiation. For a laser polarization perpendicular to this plane, Raman scatter can be detected; hence, the Raman signal can be separated from the fluorescence signal based on laser polarization. This process is not preferred because of the necessity of rotating the polarization of the excitation source; however, polarizing the laser source can increase the ratio of the Raman scatter to the fluorescence yield.

The accuracy of these separation techniques depends in part on the relative magnitudes of the signals being separated. Measurements conducted to date indicate that, at least in parts of the flame, the Raman scatter is of the same magnitude as the

fluorescence and measurements published in Ref. 3 indicate that, if a Krypton Fluoride (KrF) laser is used, the Raman signal is larger than the fluorescence for most flame conditions. In that case, signal to noise ratios for the extracted Raman signal can be expected to be comparable to those of the raw measurements.

The scope of the work detailed in this manuscript includes a description of the basis for the technique and a discussion of a set of measurements demonstrating the fact that the Raman scatter can be separated from the fluorescence based on polarization. This discussion is followed by a general outline for implementing the technique in a combustor.

Theory

Both Rayleigh and Raman scattering can be modeled classically as radiation from an induced oscillating dipole. The electric and magnetic fields of the incident radiation induce the oscillating dipole. When the induced dipole oscillates at the frequency of the incident radiation, the scattering, which is at the frequency of the incident radiation, is classified as Rayleigh. It is possible for the scattering process to occur with the irradiated molecule giving up or absorbing discrete amounts of energy. This process is referred to as Raman scattering and the scattered light is at a different frequency from the incident radiation; nevertheless, the oscillating dipole radiation model provides fundamental insight into the

directional and polarization characteristics. Using the classical oscillating dipole model, it can be seen that for scatter in a direction perpendicular to the electric field vector of the incident radiation, the electric field vector of the scattered light will be parallel to the electric field of the incident light. A more complete theoretical description of the polarization characteristics of Raman scatter arise from quantum mechanical treatments and can be described using the Stokes parameters. The Stokes parameters for Raman scatter by a diatomic molecule are given in Ref. 1 for scattering perpendicular to the incident

of conditions; however, some insight concerning expected species concentrations can be gained from equilibrium calculations. Figure 1 contains plots of the molecular species present in the flame conditions expected based on equilibrium calculations for a range of OF equivalence ratios. The LOX-rich regions would be to the right of unity in these plots and, since the injection ratio is substantially LOX rich, the conditions away from the immediate vicinity of the injectors would be LOX rich. Molecular oxygen and water are both present in substantial amounts for these conditions; however water is not a diatomic molecule and it cannot currently be used as a thermometric species. Hence molecular oxygen has been chosen as the probe species for the Raman measurements.

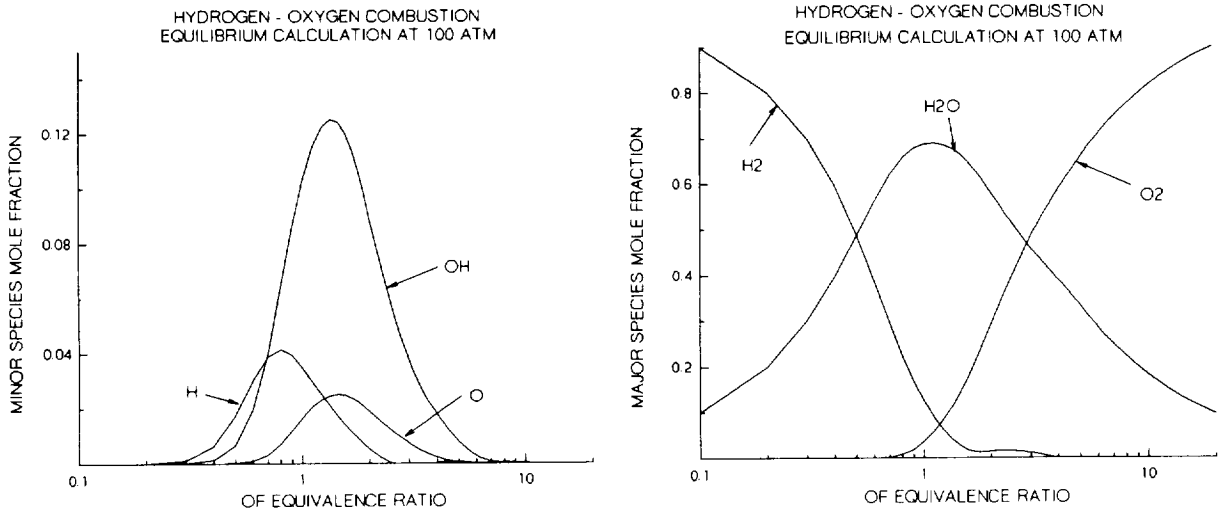

Figure 1: Species concentrations for LOX-Hydrogen combustion at 100 atm.

Experimental Setup

A schematic of the laboratory Raman scattering system is shown in Fig. 2. The

telescope consisting of a 30 cm focal length lens (first) and a 20 cm focal length lens. The beam is then focused into the control volume using a 50 cm focal length lens. The scattered signal is collected and analyzed using a double monochromator (ISA JY Ramanour). The photomultiplier tube (PMT) connected to the double monochromator is a Hamamatsu R1477. The box car integrator is from SRS and is triggered using a fiberoptic and a Hamamatsu R928 PMT. The collection optic for this work is a 60 cm focal length Parabaloid. The flame is generated using a burner equipped with hydrogen ports in a central region and oxygen ports in a circular pattern surrounding the hydrogen ports.

Results

The central idea conveyed by the results obtained thus far is a demonstration of the discrimination between the Raman signal and the fluorescence signal based on polarization. The plot shown in Fig. 3 shows spectra obtained at the wavelength expected for the Raman scatter for the oxygen molecule using the XeCl excimer laser. This demonstration measurement was conducted by collecting spectra with horizontal and vertical laser polarizations rather than by discriminating between polarization in the collection path for instrumental reasons. The signals for the two polarizations and the difference between the signals are all plotted. Each of the raw signals contains the fluorescence. The signal with the vertically polarized beam also contains the Raman and the difference between the signals is the oxygen Raman scatter.

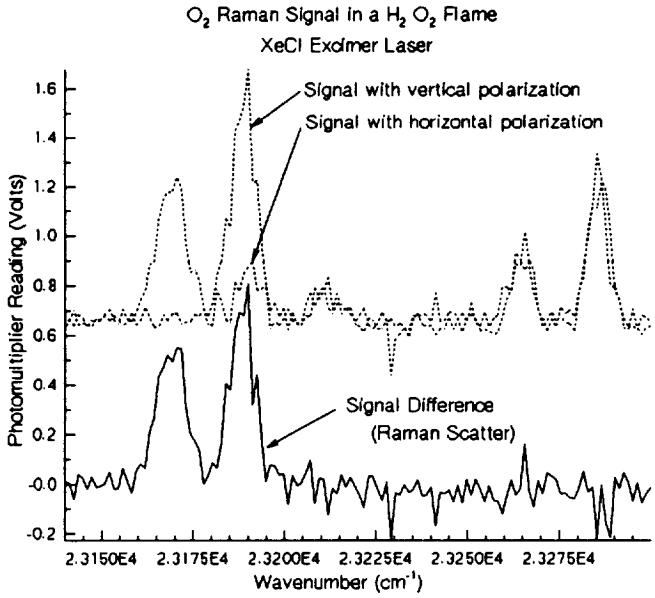

Figure 3: Raman signal separated based on laser polarization

Summary and Future Work

The measurements conducted to date have demonstrated the principle that Raman scattering signals can be separated from fluorescence signals based on polarization. The temperature and number density information will be extracted from the Raman signals

collected in the laboratory setup as a demonstration of the measurement approach. Additionally, a plan for implementing this technique in a combustor has been developed. A schematic of the optical setup proposed for the combustor measurements is shown in Fig. 4. This setup is similar to the laboratory rig with some notable exceptions. An intensified CCD array will replace the PMT to eliminate the need for scanning the monochromator and allow for single-shot measurements. A polarization separation prism will be included in the collection optics so that the Raman signal can be separated from the fluorescence without rotating the polarization of the laser beam. This too is necessary for single shot measurements. The monochromator will only pass vertically polarized light efficiently; hence a half wave plate is necessary to get the initially horizontally polarized component of the signal through the monochromator. An ellipsoidal reflecting collection optic has also been designed for use in the combustor rig.

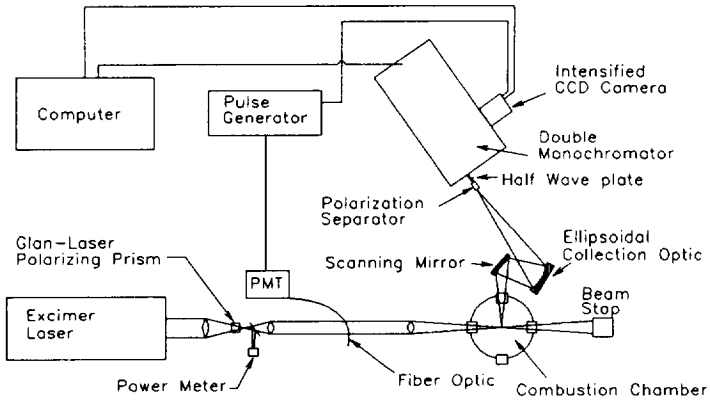

Figure 4: Schematic of Proposed Optical Setup

Acknowledgement

This document is a report on the summer faculty fellowship for Roy Hartfield; however, the author is only a small contributor offering support to the effort and suggestions concerning the polarization to an ongoing effort designed to conduct measurements of temperature and species concentration in a liquid rocket motor combustor. Chris Dobson, Richard Eskridge and Mike Lee are all principle contributors to that project.

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