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 Background Emission With Single-Shot
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DEVELOPMENT OF A TECHNIQUE FOR SEPARATING RAMAN SCATTERING SIGNALS FROM BACKGROUND EMISSION WITH SINGLE-SHOT MEASUREMENT POTENTIAL

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Abstract

A novel technique for extracting Q-branch Raman signals scattered by a diatomic species from the emission spectrum resulting from the irradiation of combustion products using a broadband excimer laser has been developed. This technique is based on the polarization characteristics of vibrational Raman scattering and can be used for both single-shot Raman extraction and time-averaged data collection. The Q-branch Raman signal has a unique set of polarization characteristics which depend on the direction of the scattering while fluorescence signals are unpolarized. For the present work, a calcite crystal is used to separate the horizontal component of a collected signal from the vertical component. The two components are then sent through a UV spectrometer and imaged onto an intensified CCD camera separately. The vertical component contains both the Raman signal and the interfering fluorescence signal. The horizontal component contains the fluorescence signal and a very weak component of the Raman signal; hence, the Raman scatter can be extracted by taking the difference between the two signals. The separation of the Raman scatter from interfering fluorescence signals is critically important to the interpretation of the Raman for cases in which a broadband ultraviolet (UV) laser is used as an excitation source in a hydrogen-oxygen flame and in all hydrocarbon flames. The present work provides a demonstration of the separation of the Raman

scatter from the fluorescence background in real time.

Introduction

Raman scattering is a powerful technique for quantitatively probing high temperature and high speed flows.¹ Raman spectroscopy is uniquely capable of providing quantitative measurements of temperature and species number density in many types of flows;^{2,3,4} however, the utility of this technique for hydrocarbon flame applications has typically been limited because of the broadband fluorescence interference. Fluorescence can also interfere with the Raman signal in clean hydrogen flames when broadband UV lasers are used as the scattering source. A solution to this problem has been demonstrated.^{5,6,7} An excellent solution to the fluorescence interference problem for many applications lies in the fact that the vibrational Q-branch Raman signal is highly polarized for 90° signal collection and the fluorescence background is essentially unpolarized. Two basic schemes are available for separating the Raman from the background. One scheme involves using a polarized laser and collecting a signal with both horizontal and vertical laser polarizations separately. For vertical laser polarization, the signal with the vertical polarization will contain both the Raman and the fluorescence while the signal with the horizontal polarization will contain only the fluorescence. This is the approach followed in Refs. 5 and 6. The second scheme involves polarization discrimination on the collection side of the optical setup. For vertical laser polarization, the scattered Q-branch Raman signal will be vertically polarized; hence the two

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polarizations can be collected separately and the difference between the two is the Raman signal. This approach has been used for the work found herein and has the advantage of allowing the data to be collected from the same laser shot(s). This makes it possible to collect quantitative Raman data with single shot resolution in conditions where interference cannot otherwise be eliminated.

Experimental Setup

The experimental arrangement used to demonstrate this approach consists of a Heinken burner, a Lumonics Broadband Krypton Fluoride excimer laser (248.6 nm), a McPherson 1 m UV spectrometer, a computer controlled Princeton Instruments intensified CCD camera system, a pulse generator and a calcite crystal used for separating polarizations. The schematic found in Fig. 1 shows

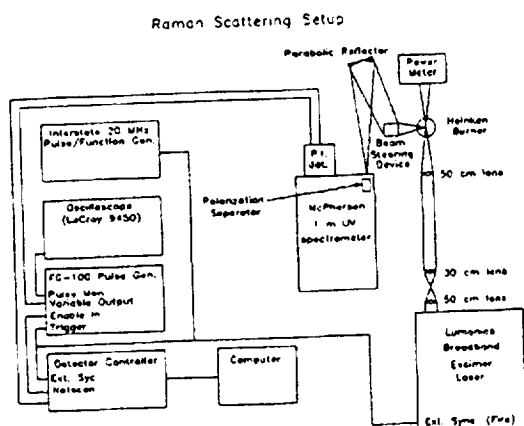


Figure 1: Experimental Setup

the experimental setup including appropriate optics for focusing the laser beam over the burner and collecting the resulting signal. The schematic includes the essential electronic connections needed for proper operation of the camera in the gated, synchronized mode. The pulse/function generator was used to fire the laser on the positive going pulse and the camera on the negative going pulse. This allowed the function generator pulse width to be used as a delay. The 15 ns. laser pulse was centered on a 100 ns. camera gate using a pulse monitor from the

FG-100 pulse generator and a photomultiplier (not shown) looking at stray light from the laser beam. The key element which is unique for this setup is the polarization separator. This crystal separates a beam of unpolarized incoming light into horizontal and vertical components separated by 4 mm. The crystal was placed just inside the slit of the spectrometer. A mask approximately 3mm in diameter was placed in from of the slit resulting in a pattern on the detector composed of two strips of light, each containing one spatial dimension and one spectral dimension.

Results

The immediate form of the data is electronic images of the horizontally and vertically polarized components of the collected scatter. The image shown in Fig. 2 represents a sample of the raw data.

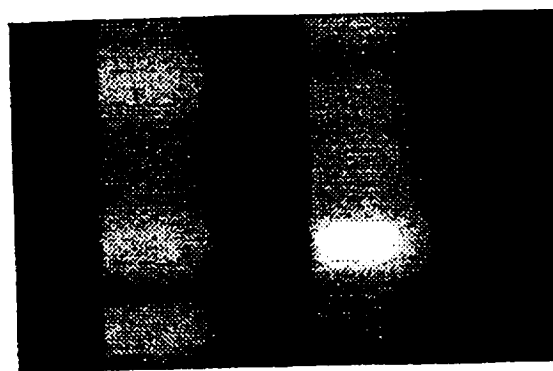


Figure 2: Image of spectrally resolved scattering signal along a line separated based on polarization

The vertically polarized light appears on the left band and the horizontally polarized light appears on the right band. The spectral direction is along the vertical axis with wavelength increasing from bottom to top and the horizontal direction represents distance along a linear probe volume. This particular image was collected in a lean flame ($OF = 4$) with the spectrometer set at 269.8 nm. (This wavelength corresponds to the approximate center of the image.) The horizontally polarized band is generally brighter because the spectrometer passes horizontally

polarized light preferentially; however, some bands appear on the vertically polarized section which are not present on the horizontally polarized image. These areas represent Raman scatter. The two polarizations are normalized for collection efficiency and then their difference is taken to extract the Raman spectrum. This process is illustrated by the rough plots of data shown in Figs. 3 and 4.

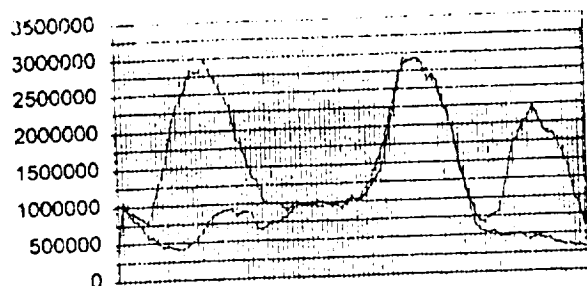


Figure 3: Plot comparing vertically and horizontally polarized components of induced scatter

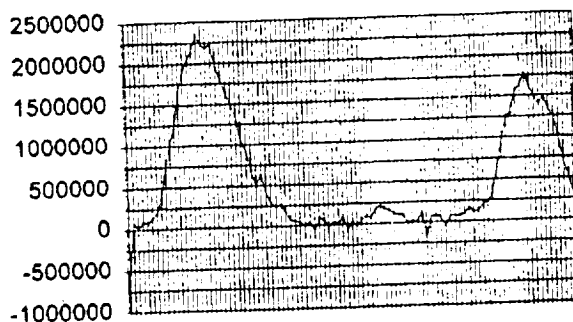


Figure 4: Difference between horizontal and vertically polarized components (Raman Signals for N_2 (left) and O_2 (right))

The data for figs. 3 and 4 was taken from a 500 shot average image and then binned in the spatial

direction for maximum signal strength. The spatial averaging is possible because the linear probe volume segment for this image is contained entirely in the core of the flame where the conditions are uniform on a time-averaged basis.

An additional data set showing the vertically polarized signal, the horizontally polarized signal and the extracted O_2 and N_2 Raman signals is shown in the plot in Fig. 5. This data was also collected in a

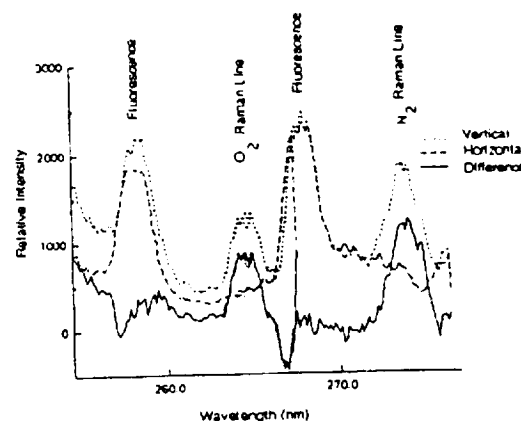


Figure 5: Horizontal and Vertical signals in a lean flame

lean hydrogen-air flame with an OF ratio of 4 but in a slightly different area of the flame. Again, this data demonstrates an effective isolation method for the Raman data.

An additional issue to be addressed in the development of the technique to separate the Raman from the background is the polarization of fluorescence signals depending, perhaps, on the degree of predissociation for the absorbing molecule and the fluorescence lifetimes. This approach assumes that the fluorescence is completely unpolarized and that must be verified. As a preliminary verification, some data was collected in a propane-air flame with a narrowband Lambda Physik excimer laser and a slightly different spectrometer setup. This data is presented in Figs. 6-9. These images show the vertically polarized signal

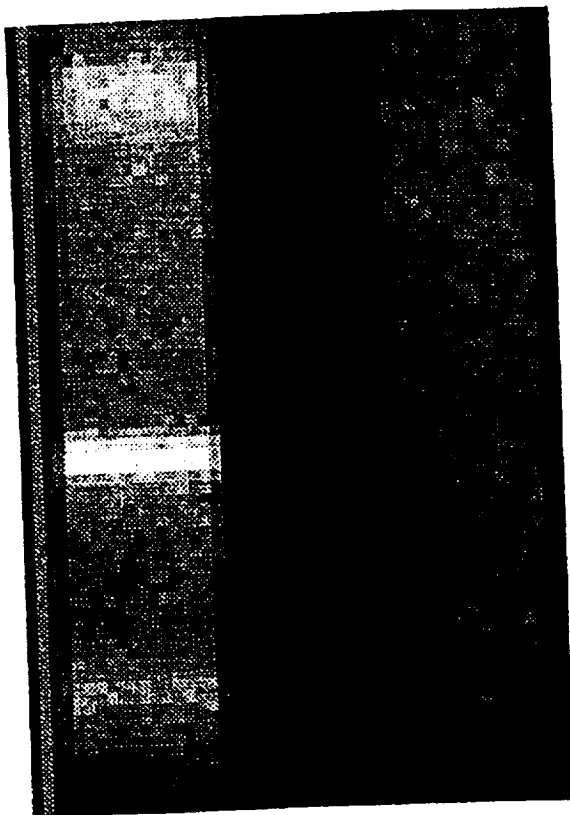


Figure 6: Polarization based separated signals from narrowband excitation of a slightly lean propane flame with laser detuned from absorption transitions

on the left and the horizontally polarized signal on the right. For the image shown in Fig. 6, the oxygen, nitrogen and water Raman signals are visible from bottom to top. This is a lean, near-stoichiometric flame and the laser is tuned off-resonance of either OH or O_2 absorption transitions. There is definitely some laser-induced broadband fluorescence interference which is present on both the left and right side of the image. Figure 7 is data for the same conditions as those for Fig. 6 but with the laser tuned to a weak OH transition. The three fluorescence emission lines (P,Q, and R branches) for that transition are apparent for both polarizations around the water Raman signal. Figure 8 is for the same conditions again but with the laser tuned to a stronger OH transition.

The data presented in Fig. 9 is the same flame but the laser is tuned onto an O_2 absorption

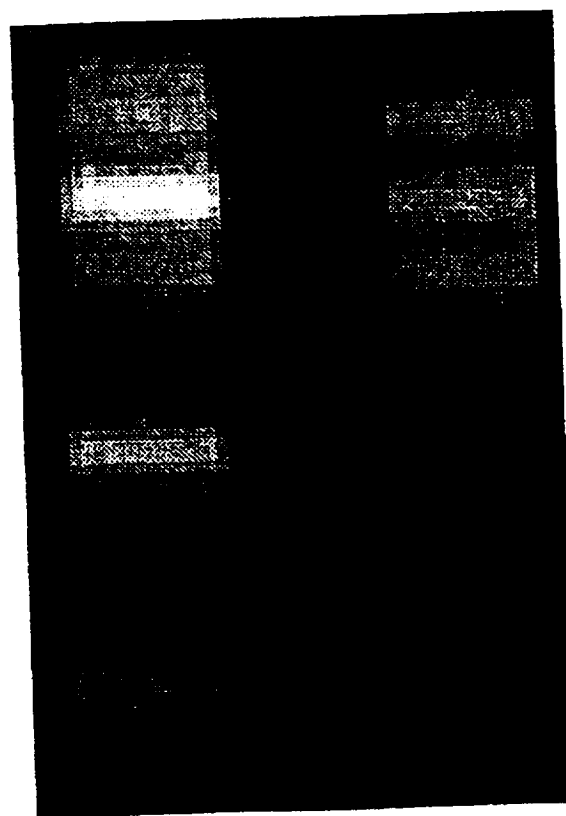


Figure 7: Polarization based separated signals for a slightly lean propane flame using narrowband laser excitation with laser tuned to a weak OH absorption transition

line. The resulting O_2 fluorescence is visible for both polarizations, with three bands located near the O_2 Raman line, just on the long wavelength side of the N_2 Raman line, and just on the long wavelength side of the water Raman line.

Additional analysis of this data is necessary to quantify the degree of polarization for the O_2 and OH fluorescence signals but an initial look at this data indicates that the degree of polarization for these two signals is small.

The procedure for measuring temperature with the data available in this report is to collect data in a known flame condition to determine the correlation between the Raman signal and the number density, conduct Raman measurements in and unknown flow field at a known pressure, calculate the number density from the Raman signals and then

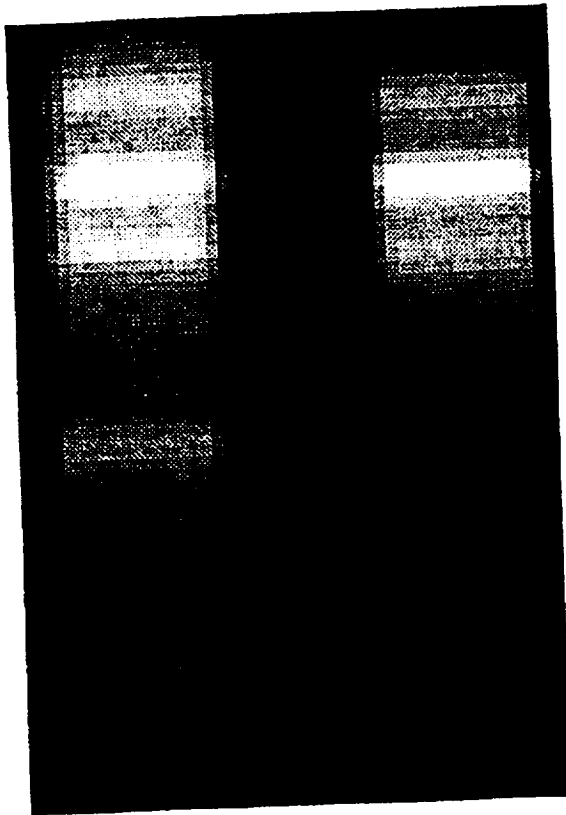


Figure 8: Polarization based separated signals using narrowband excitation in a slightly lean propane flame with laser tuned to a stronger OH absorption transition

calculate the temperature using ideal gas. It should be pointed out that, with a narrowband laser, the temperature can be determined directly from the shape of the Raman signal and then number densities (and partial pressures) can be determined from the total Raman signal strength. (This approach will be proposed for future hydrocarbon technique development.

Summary and Future Work

A novel technique for extracting Raman signals from interference has been demonstrated. This technique offers single shot measurement potential, quantitative use of Raman signals obtained using broadband UV lasers and the possibility of obtaining quantitative Raman data in hydrocarbon flames. Future development of the technique should include a completion of the verification of the

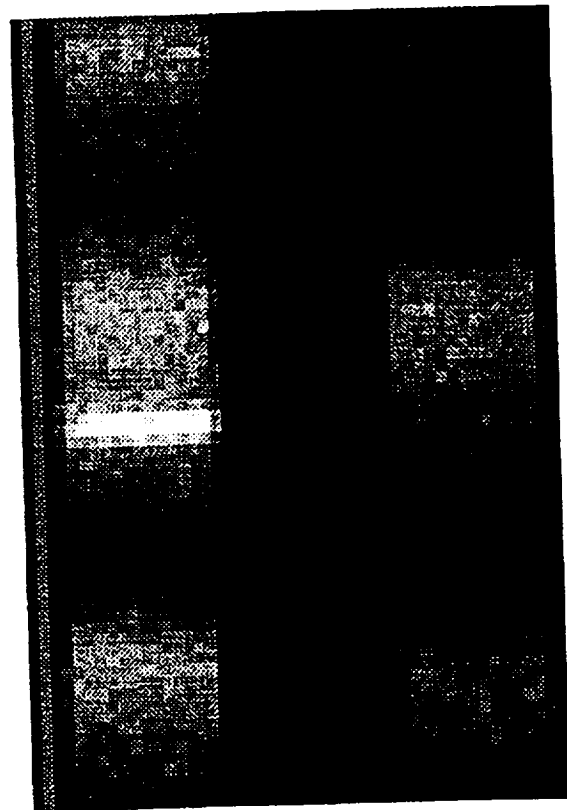


Figure 9: Polarization based separated signals in a slightly lean propane flame with narrowband laser excitation with laser tuned to an Oxygen transition.

polarization characteristics of the interfering fluorescence signals and calculations of the temperature in a variety of combustion flow field environments.

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