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LASER SPECTROSCOPY OF PROPELLANT-LIKE FLAMES

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The laser-based techniques Coherent Anti-Stokes Raman Spectroscopy (CARS) and Spontaneous Raman Scattering have been applied to studies of the detailed chemistry of premixed flames. The status of this research effort and its future goals in gun propulsion research are discussed.

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I. INTRODUCTION

During the ignition and combustion of gun propellants there is a complex interplay between chemical and physical processes which determines the performance of these materials as well as the system in which they are used. It is the goal of the present work to develop a detailed understanding of the controlling processes of some typical gun propellants through a program of measurements on model flame systems and computer simulation to extend the results to conditions encountered during the interior ballistic cycle. An important part of this program is the revolutionary tools based on lasers and light detection techniques which have become available for active probing of combustion.[1] The development and application of these tools to such problems as air pollution and energy conversion efficiency has led to a broad scientific base in the academic and industrial communities. This base provides substantial technical support to the present program. Although these techniques are not necessarily well-suited for application to probing the surface of a burning solid, it is possible to understand the gas phase chemistry immediately above the surface; it is expected that the heat release in this region is important in many situations. The extension and application of these results to ignition and early combustion of gun propellants should be straightforward.

The major stable decomposition products of HMX and RDX are[2] HCHO, HCN, N_2O, and NO_2. On the basis of kinetics studies[3] it is highly probable that the $HCHO/NO_2$ flame system releases heat faster than any other combination from this list. Thus, this flame may be dominant in providing heat feedback to the propellant surface and hence be the most important gas phase contribution to the propellant burning rate. However, it is important to note that the kinetics of combinations of these species have not been studied, and behavior in a flame may be different from that expected from simple kinetic arguments. As this work progresses, flames will be studied in the following order. In order to validate the models and techniques, a methane/air system is being studied as the first system. Further work will also use methane in place of formaldehyde, which is extremely difficult to work with as a pure monomer. Following methane/air will be methane/N_2O, hydrogen/N_2O, hydrogen/NO_2, carbon monoxide/hydrogen/NO_2, formaldehyde/NO_2, and then mixtures of other fuels and oxidizers.

[1a] *A.C. Eckbreth, P.A. Bonczyk, and J.F. Verdieck, "Laser Raman and Fluorescence Techniques for Practical Combustion Diagnostics," App. Spec. Rev., Vol. 13, p. 15, 1977.*

[1b] *Laser Probes for Combustion Chemistry, D. Crosley, Ed., ACS Symp. Series, No. 134, 1980.*

[2] *C.U. Morgan and R.A. Beyer, "ESR and IR Spectroscopic Studies of HMX and RDX Thermal Decomposition," Proceedings of 15 JANNAF Combustion Meeting, CPIA Publication 297, 1978.*

[3] *R.A. Fifer and H.E. Holmes, "Kinetics of the HCN + NO_2 Reaction Behind Shock Waves," J. Phys. Chem., Vol. 86, p. 2935, 1982.*

In addition to the flame diagnostics a considerable effort must be made in computer simulation of these flames in order to interpret the measurements and to extend them to propellant combustion conditions. Substantial progress has been made in this area with development of premixed flame codes;[4] quality data from flame measurements are required to progress further.

Measurements on laboratory flame systems can be divided into two areas: (1) those which detect major species, i.e., those present in concentrations of 0.1 percent or more, and (2) those techniques which are much more sensitive and detect radical species which are important even though they are typically present at a parts per million level. The primary technique used here for radical detection is laser induced fluorescence (LIF); the application of LIF to several flame systems is detailed elsewhere.[5] Major species concentrations are measured using two Raman techniques: spontaneous Raman scattering and Coherent Anti-Stokes Raman Spectroscopy (CARS). Because of its relative ease of application and interpretation, spontaneous Raman spectroscopy is the technique of choice for clean, stable systems. CARS has been well demonstrated[6] to be an excellent probe for highly turbulent, sooty, or otherwise hostile flame systems. All of these techniques also are capable of accurate temperature measurements, as is seen below and in the following paper.[5]

II. SPONTANEOUS RAMAN DIAGNOSTICS

The experimental apparatus used for spontaneous Raman measurements in these flames has two important features: the use of a continuous wave argon ion laser in an intracavity mode to provide a useable power of about 70 watts, and multichannel vidicon detection. A schematic of the apparatus is shown in Figure 1. The laser cavity is extended as shown by removing the standard output mirror and adding high reflectivity mirrors of 1.0 and 0.3 m radius of curvature to provide an intracavity beam waist of about 100μm. A curved knife edge burner[7] is situated at this laser beam waist. The scattered light is collected by an f/1.3 lens through the burnt gas region of the flame and imaged onto the slits of a 0.25 m spectrometer. The dispersed light is detected by a silicon intensified target vidicon; the detector signals are digitized and stored by a dedicated PDP 11/34 laboratory computer using

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[4] *J.M. Heimerl, "A Contribution to the Flat Flame Olympics: Problem B," ARBRL-TR-02416, 1982 (AD A119401).*

[5] *W.R. Anderson, A.J. Kotlar, L.J. Decker, and S.W. Bunte, "Flame Radical Measurements Using Laser Excited Fluorescence," Proceedings of 19th JANNAF Combustion Meeting, CPIA Publication No. 366, p. 145, 1982.*

[6] *R.J. Hall and A.C. Eckbreth, "Coherent Anti-Stokes Raman Spectroscopy: Application to Combustion Diagnostics," Laser Applications, Vol. V, R.K. Erf, ed., 1981.*

[7] *R.A. Beyer and M.A. DeWilde, "Simple Burner for Laser Probing of Flames," Rev. Sci. Instrum., Vol. 53, p. 103, 1982.*

extensive software developed in this laboratory.[8] This data aquisition system also allows the computer control of burner position and spectrometer wavelength for automatic data aquisition.

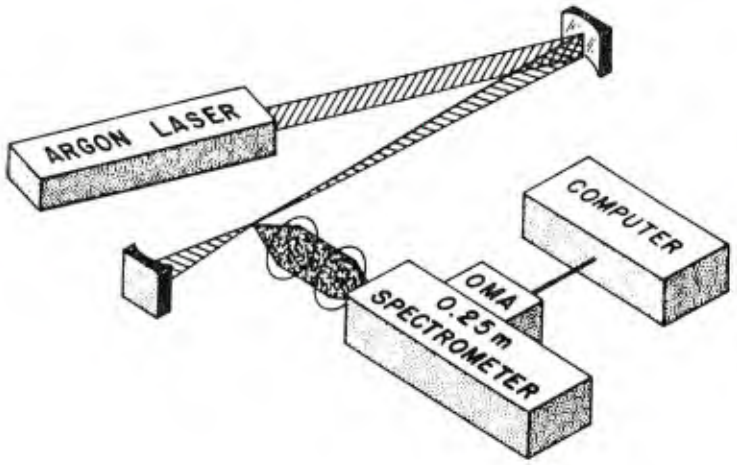

Figure 1. Schematic of the Intracavity Raman Apparatus

Two typical spectra are shown in Figure 2. The wavelength coverage is sufficiently broad to obtain most major species at two wavelength settings; moreover, resolution is sufficiently good to obtain accurate temperatures from nitrogen Stokes Q-branch rotational-vibrational spectra, as shown in Figure 3. These temperatures are obtained from a multiparameter least squares fit to the data. As an example of the quality of data possible with this system, the Stokes Q-branch spectrum of H_2 present at about the 1.3 percent level in a slightly rich methane/air flame is shown in Figure 4.

A series of profiles of temperature and some of the major species in a slightly rich (ϕ = 1.4) methane air flame is shown in Figure 5. Note that the lines are for clarity only. In these data one can possibly observe the effects of air entrainment late in the reaction zone causing oxidation of the H_2 and CO. These data demonstrate the quality of data available from this system.

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[8] *M.A. DeWilde, "OMADAS, An Automatic Data Acquisition and Analysis Program for the PARC OMA," to be published.*

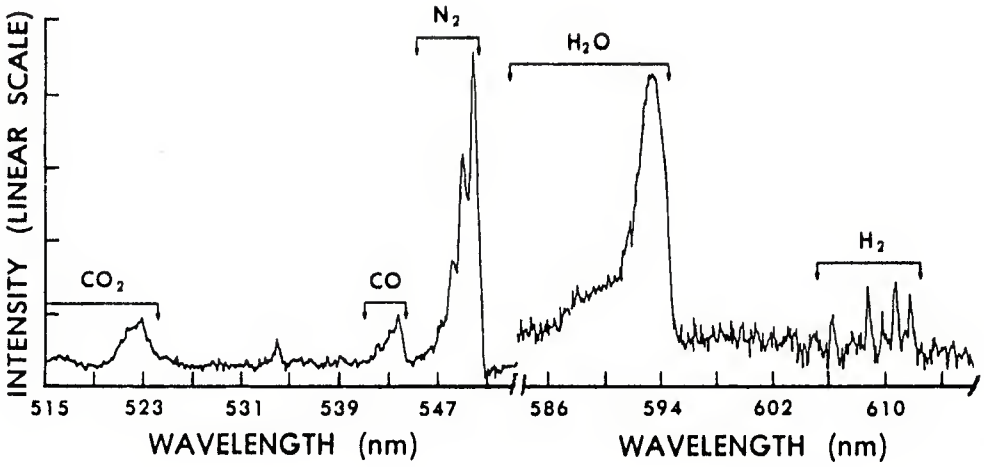

Figure 2. Two Typical Raman Spectra from a Methane/Nitrous Oxide Flame

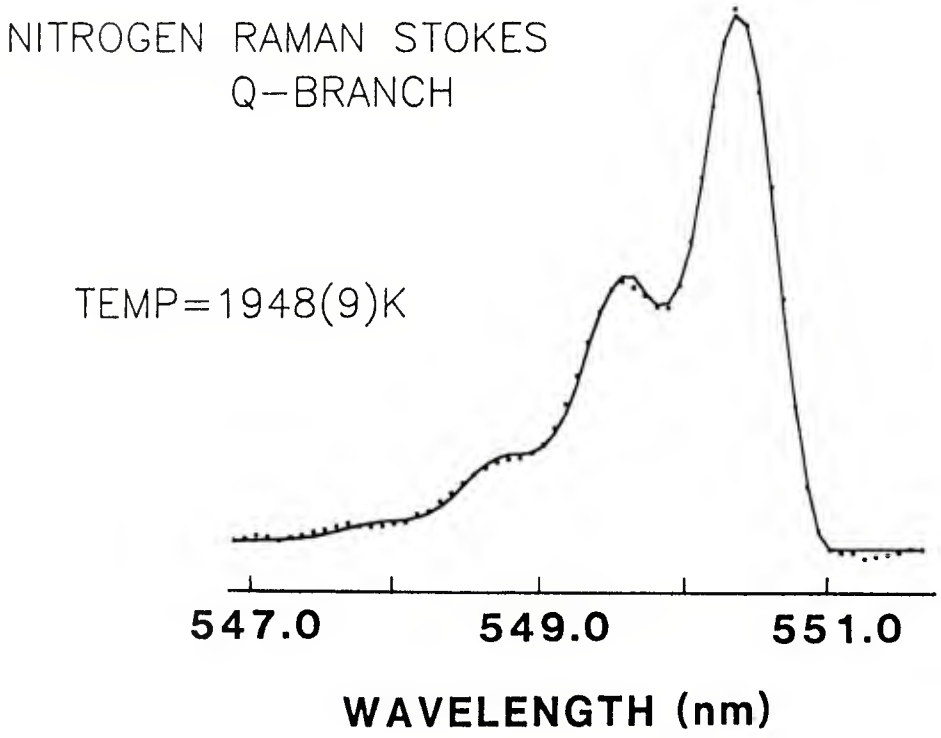

Figure 3. Nitrogen Stokes Q-Branch Raman Spectrum with Data (•) and Computer Fit.

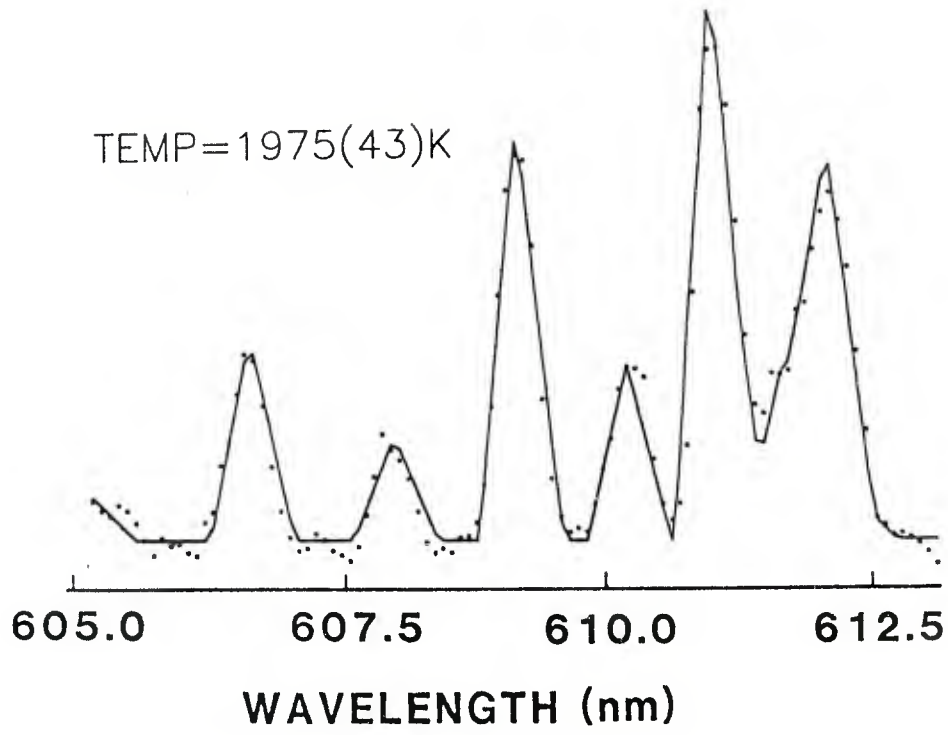

Figure 4. Hydrogen Q-Branch Raman Spectrum Early in a Slightly Rich Methane/Air Flame Where T ≈ 850K.

III. COHERENT ANTI-STOKES RAMAN SPECTROSCOPY

A limited amount of research has been done utilizing CARS as a diagnostic for major species in steady state flames with a goal of applying it to transient combustion processes. The system consists of a high power Nd:YAG laser frequency doubled to produce ~500 mJ/pulse at 532 nm. Part of this output is used to pump a broadband dye laser oscillator and amplifier. The dye laser output and remaining 532 nm radiation are then used to generate a CARS signal using either a colinear or BOXCARS configuration. The signal is separated from the laser beams using a prism and beam stops, dispersed by a 0.3 m monochromator, and detected with a vidicon tube as in the spontaneous Raman studies. Much of the development work of CARS thermometry has been with the nitrogen Q-branch spectrum. A typical series of these spectra in a methane/N_2O flame is shown in Figure 6. Also shown in the Figure are the computer fits from an active multiparameter computer fit to the CARS data. A second molecule of interest locally for thermometry, at lower temperatures and in rich flames, is hydrogen. A typical H_2 CARS spectrum obtained in this work is shown in Figure 7. Note that a substantial nonresonant susceptibility contribution underlies the signals. As is mentioned below, this technique is currently being pursued as a diagnostic for application to gun propulsion measurements.

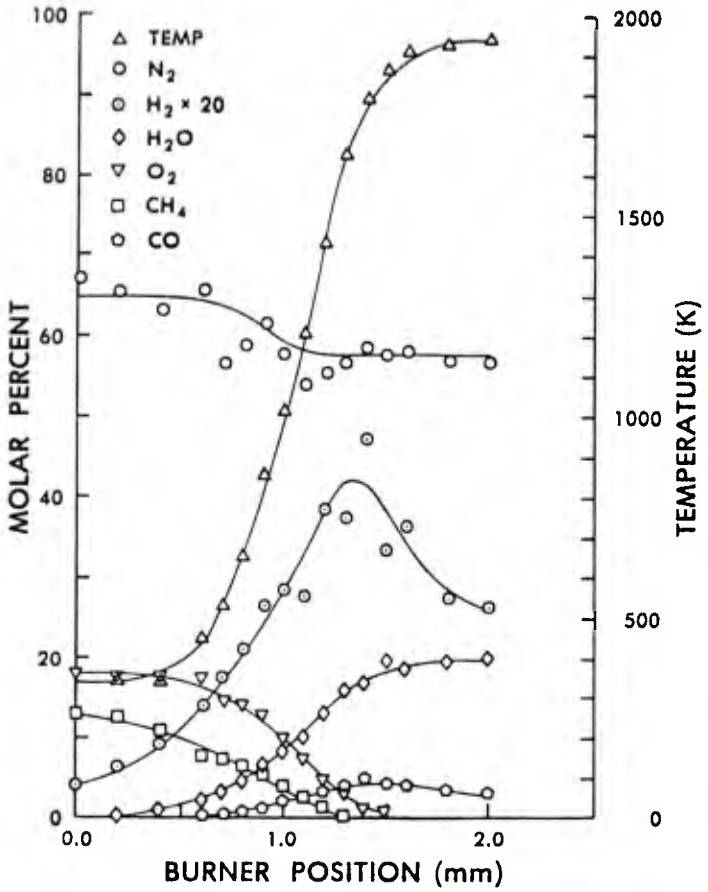

Figure 5. Profiles of Temperature and Major Species Through the Primary Reaction Zone of a Methane/Air Flame

IV. FUTURE DIRECTIONS

In addition to pursuing the understanding of the various flames as mentioned above, efforts are now being made to apply these techniques to less well-behaved real systems. The first such application will be utilizing CARS thermometry to measure gun muzzle exhaust temperatures and temperatures above burning propellants in a windowed strand burner. As these efforts progress, it is expected that CARS will also be developed to measure species concentrations of major species in these environments.

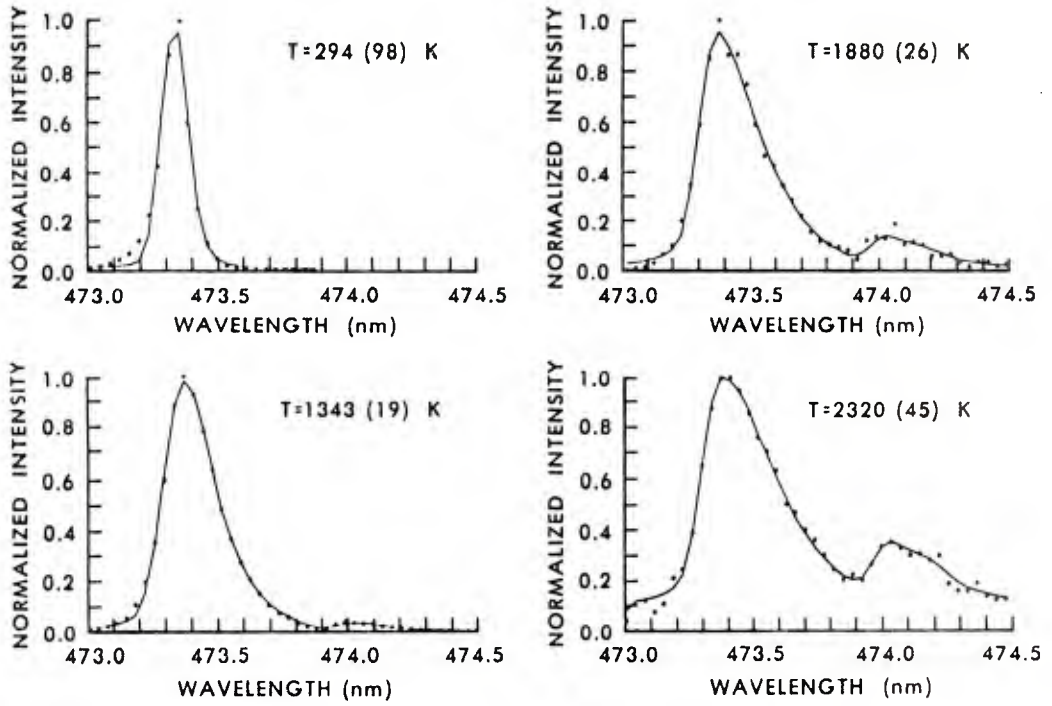

Figure 6. Nitrogen CARS Spectra from Four Positions in a Methane/Nitrous Oxide Flame Showing Data (•) and Computer Fit

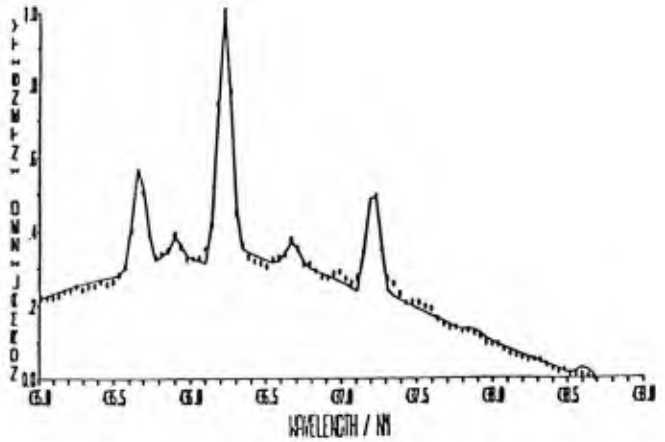

Figure 7. Hydrogen CARS Spectra From a Rich Methane/Nitrous Oxide Flame Showing Data (•) and Computer Fit

The second major area of application involves the use of laser induced fluorescence (LIF) to study the radical species in burning propellants as well as their role in muzzle flash. An important part of this program is the further development of the two-dimensional imaging of LIF as previously demonstrated[9] on simple systems. Both point measurement LIF and the 2-dimensional technique should provide key information about radical concentrations and distributions in these situations.

Overall, the combination of accurate temperature measurements, radical species distributions, and computer simulation of these systems should provide us with the key to control of these processes so that we can enhance performance of future systems.

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[9] *M.J. Dyer and D.R. Crosley, "Two Dimensional Imaging of OH Laser-Induced Fluorescence in a Flame," Opt. Letter, Vol. 7, p. 382, 1982.*

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2. Does this report satisfy a need? (Comment on purpose, related project, or other area of interest for which report will be used.)

_____

_____

_____

3. How, specifically, is the report being used? (Information source, design data or procedure, management procedure, source of ideas, etc.) _____

_____

_____

4. Has the information in this report led to any quantitative savings as far as man-hours/contract dollars saved, operating costs avoided, efficiencies achieved, etc.? If so, please elaborate.

_____

_____

5. General Comments (Indicate what you think should be changed to make this report and future reports of this type more responsive to your needs, more usable, improve readability, etc.) _____

_____

_____

_____

6. If you would like to be contacted by the personnel who prepared this report to raise specific questions or discuss the topic, please fill in the following information.

Name: _____

Telephone Number: _____

Organization Address: _____

_____

_____