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Stokes Raman Scattering**

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MEASUREMENTS OF TEMPERATURE AND CO₂ CONCENTRATION BY DUAL-PUMP COHERENT ANTI-STOKES RAMAN SCATTERING

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ABSTRACT

Measurements of temperature and CO₂ concentration using dual-pump coherent anti-Stokes Raman scattering (CARS) are described. The measurements were performed in laboratory flames, in a room temperature gas cell, and on an engine test stand at the Air Force Research Laboratory at Wright-Patterson Air Force Base. A modeless dye laser, a single-mode Nd:YAG laser, and an unintensified back-illuminated charge-coupled device (CCD) digital camera were used for these measurements. The CARS measurements were performed on a single-laser-shot basis. The standard deviation of the temperatures and CO₂ mole fractions determined from single-shot dual-pump CARS spectra in steady laminar flames were approximately 2% and 10% of the mean values of approximately 2000 K and 0.10, respectively. The application of dual-pump CARS for single-shot measurements in a swirl-stabilized combustor fueled with JP-8 was also demonstrated.

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INTRODUCTION

Coherent anti-Stokes Raman scattering (CARS) has been applied widely for the measurement of temperature in a variety of practical combustors [Eckbreth (1996), Greenhalgh et al. (1983), Eckbreth et al. (1984), Goss et al. (1983), Alden and Wallin (1985), Bengtsson et al. (1994), Switzer et al. (1994)]. However, the use of CARS for measurements of species concentrations in combustor flowfields has been much less common [Eckbreth et al. (1984), Eckbreth et al. (1988), Antcliff and Jarrett (1987), Green et al. (1998)]. Dual-pump CARS, demonstrated originally by Lucht (1987), is a technique where CARS spectra from two species are generated simultaneously and with nearly the same frequency. In dual-pump CARS, a narrowband, tunable dye laser is used to provide the second pump beam for the CARS process; in normal, two-color CARS, the 532-nm second-harmonic Nd:YAG laser radiation is used for both pump beams. The use of dual-pump CARS for the simultaneous measurement of N_2/O_2 (Lucht, 1987; Hancock et al., 1997a), N_2/H_2 (Schauer, 1998), N_2/CH_2 (Green et al., 1998), and N_2/CO_2 (Bruggeman, 1995) has been demonstrated. In addition, simultaneous measurements of pressure and temperature have been performed by detecting simultaneously the vibrational and pure rotational transitions of the nitrogen molecule (Foglesong et al., 1998).

Unlike other multispecies CARS techniques such as dual-Stokes and dual-broadband CARS (Eckbreth, 1996), dual-pump CARS signals from the two species of interest are generated by the same three laser beams. Thus, the CARS spectra for the two species have nearly the same frequency, simplifying greatly the detection of the dispersed CARS spectra and essentially eliminating potential errors caused by variations in signal transmission

or detector efficiency with signal frequency. In addition, a high degree of correlation between the two signals on single laser shots has been demonstrated, and the relative signal levels from the two species do not depend on system parameters such as the pulse energies of each of the three laser beams.

A series of dual-pump CARS measurements of CO_2 and N_2 was performed on flames and combustor test facilities at the Air Force Research Laboratory at Wright-Patterson Air Force Base. The objective of the experiments was to demonstrate the potential for single-laser-shot measurements of temperature and the concentration of important product species such as CO_2 in engine test facilities. Such single-shot measurements will be very useful for evaluating the combustion performance and efficiency of experimental combustors. The CARS spectra were analyzed using the Sandia CARSFIT code (Palmer, 1989) and experimental and theoretical CO_2 CARS spectra are compared.

EXPERIMENTAL SYSTEM

The dual-pump CARS system for CO_2/N_2 is depicted schematically in Fig. 1. The pump source for the measurements was a Spectra-Physics GCR injection-seeded, Q-switched Nd:YAG laser with a repetition rate of 10 Hz and a peak pulse energy at 532 nm of 1.0 J. The 532-nm output of the Nd:YAG laser was used to pump both a narrowband and a broadband dye laser. The narrowband dye laser was a Continuum ND6000 with a bandwidth of 0.08 cm^{-1} . The wavelength of the narrowband dye laser was 560 nm. The broadband dye laser was a modeless design with side-pumped Bethune cells for both the "oscillator" and amplifier (Hahn et al., 1997). The spectral output of the broadband dye laser was centered at 607 nm with a bandwidth of greater than 100 cm^{-1} .

The CARS signal was generated using the three-dimensionally phase-matched arrangement shown in Fig. 2 (folded BOXCARS), and focused onto the entrance slit of a SPEX 1-meter spectrometer. The spatial resolution of the CARS probe is estimated to be approximately 2 mm, the interaction length over which the CARS signal is generated. The wavelength of the N₂ and CO₂ CARS signals was approximately 496 nm. The wavelength of the nitrogen CARS signal was adjusted by tuning the wavelength of the narrowband dye laser. The wavelength of the CO₂ CARS signal did not vary as the narrowband dye laser wavelength was tuned, so that the relative spectral positions of the N₂ and CO₂ CARS signals could be adjusted. The CARS signal was detected using a 16-bit back-illuminated PixelVision SpectraVideo CCD camera with a 165 × 1100 array of pixels (each pixel 24- μ m square) at the exit plane of the spectrometer. To acquire single-laser-shot spectra at the laser repetition rate of 10 Hz, charge within each of the 1100 columns of the CCD array was accumulated in the serial register before readout; the collected charge was then digitized at a relatively low rate of 50 kpix/s. Even at this low readout rate, this camera is capable of achieving framing rates of $\sim 38 \text{ s}^{-1}$ (reading out all 1100 elements); this is aided by a modification of the parallel-shift drivers to accommodate a 24- μ s shift time. This thermo-electrically cooled camera (CCD temperature controlled at 245 K) also exhibited very low read noise, approximately 1.5 counts out of 65,536, and dark current, approximately 2 counts/sec, while exhibiting high quantum efficiency, 80% at 500 nm. With this SITE CCD chip, individual light-sensitive pixels have a full-well potential of about 300,000 electrons, while that for the serial-register pixels is greater than 500,000 e⁻; the gain of the digitizer was set to about 8 e⁻/DN, so

that the digitizer maximum corresponded to the serial register full-well capacity. Peak CARS signals from the McKenna-burner flame ($T \approx 1900 \text{ K}$) were typically ~ 5000 counts (equaling $\sim 40,000$ photo-electrons). Note that while the camera incorporates an integral Uniblitz 35-mm-aperture shutter, the shutter was left open during data collection, as it cannot operate at 10 Hz. Nonetheless, background flame emission—even in the case of the model gas-turbine combustor—was not sufficiently large to warrant additional shuttering (apart from the readout process).

The camera and controller were linked to a personal computer (PC) with the PixelVision Lynx PCI serial card, which provides a fiber-optic link between the camera and the PC. With the fiber-optic communication, the PC can be located remotely relative to the test article, a significant advantage for operation in large-scale facilities. The PixelView (version 3.1) software package was employed to control the camera and collect CARS spectra. For each measurement (typically composed of 500 to 1000 individual single-laser-pulse measurements), the spectra were stored first in computer RAM and then written to the hard drive in a movie format, which included a collection-time tag for individual spectra. Afterwards, each spectrum was corrected for background signal (consisting primarily of the CCD offset) and for the spectral intensity distribution of the broadband dye laser output, using a nonresonant CARS spectrum generated with argon.

The system was optimized and measurements were performed in a laminar propane/air flat-flame burner and in a gas cell containing CO₂/N₂ mixtures. The CARS system was then moved to the engine test stand and realigned to perform measurements in the exhaust region of a swirl-stabilized combustor fueled with JP-8.

A considerable quantity of single-laser-shot CARS data from (1) the propane/air flame, (2) a hydrogen/air flame seeded with CO₂, (3) the gas cell, and (4) the swirl-stabilized combustor was acquired. To aid in the analysis of this data, the CARS code was modified to allow batch processing of single-laser-shot spectra, greatly accelerating the processing of the data (Foglesong, 1999).

The laminar flame and gas cell measurements were performed to assess the accuracy and precision of the dual-pump CARS measurements. Both the laminar flat flame and the gas cell provide steady, well-characterized conditions for testing the single-laser-shot performance of the dual-pump CARS system. In addition, the CO₂ CARS model in the Sandia CARS code is still under development, and the comparison of the experimental data acquired in these experiments with calculated results from the Sandia CARS code is of great interest.

EXPERIMENTAL RESULTS

CARS Measurements in Laboratory Flames

Some typical single-laser-shot spectra acquired in a stoichiometric propane/air flame stabilized on a McKenna burner and the associated theoretical fits from the Sandia CARSFIT code are shown in Fig. 3. The wavelength of the narrowband dye laser that provided the second pump beam was adjusted so that the most prominent feature of the CO₂ Raman spectrum, the 00⁰0-10⁰0 band, was positioned close to the nitrogen bandhead. As shown in Fig. 3, the theoretical fits to the single-shot spectra are excellent and the best-fit value of the CO₂ concentration is very close to the expected value of 0.11.

For the stoichiometric propane/air flames investigated, the temperature is expected to be just under 2000 K (based on previous measurements in this burner), and

the equilibrium mole fraction of CO₂ is calculated to be 0.11 in the post-flame region. Histograms of temperature and CO₂ mole fraction determined from 500 single laser shots in the laminar propane/air flame are shown in Figs. 4 and 5, respectively. The pdfs contain 480 points, and there were 20 spectra where the CARSFIT code returned the initial values at the end of the least-squares fitting procedure. We have not examined these 20 spectra in detail to determine why the fitting procedure failed, although the quality of the spectra were not noticeably different. The mean and rms standard deviation of the temperature distribution are 1988 K and 42 K, respectively. The mean and rms standard deviation of the CO₂ mole fraction distribution are 0.097 and 0.013, respectively. The measured mean CO₂ mole fraction is slightly lower than the expected value of 0.11, probably because some improvement is needed in the CO₂ spectral model in the Sandia CARS code. The sensitivity of the CARS measurement of CO₂ is affected significantly because the rotational constants in the 00⁰0 and 10⁰0 vibrational bands are nearly equal. Consequently, the Q-branch rotational transitions overlap and the CARS signal is enhanced greatly by this overlap and by significant collisional narrowing effects.

Dual-pump CARS measurements were also performed in steady propane/air/CO₂ flames stabilized on a McKenna burner. The flowrates of propane and air were held constant and the equivalence ratio was therefore constant; the flames were slightly fuel-lean. The CO₂ flowrate was varied to obtain CARS spectra at different CO₂ mole fractions. Averaged CARS spectra from two of these flames are shown in Fig. 6. The experimental results are in good agreement with theory, although the best-fit CO₂ mole fraction is slightly lower than the value expected based on the

known flowrates, as was the case for the measurements shown in Fig. 5.

Measurements were performed in near-adiabatic hydrogen/air/CO₂ flames stabilized on a Hencken burner to obtain high-quality dual-pump CO₂/N₂ CARS spectra over a wide range of temperatures (Hancock et al., 1997b). The flowrate of the CO₂ was held constant while the hydrogen flowrate and, consequently, the temperature was varied over a wide range. Because of the near-adiabatic nature of the Hencken burner the measured CARS temperatures can be compared with calculated adiabatic equilibrium temperatures. As can be seen from the spectra shown in Fig. 7, the spectra were acquired with very good signal-to-noise ratios. However, upon analyzing the spectra, the CO₂ line corresponding to the 00⁰0-10⁰0 band is much too strong for the known concentration in all of the spectra. Curiously, there is excellent agreement between theory and experiment for the other CO₂ spectral features. There is nothing in the nonresonant spectrum acquired before and after the measurements to suggest that the broadband dye laser spectrum was any different during the data acquisition. The data were analyzed several months after the experiment and it has not yet been possible to repeat the measurements.

The temperatures determined from a least squares fit of the nitrogen CARS spectrum and calculated using the NASA Lewis chemical equilibrium code (Gordon and McBride, 1976) are in excellent agreement, as shown in Fig. 8. The CARS temperature is slightly lower than the adiabatic equilibrium temperature for equivalence ratios below 0.5, and slightly higher for the richest equivalence ratio of 1.08. For the rich equivalence ratio, this disagreement may be the result of oxygen from the surrounding air penetrating to the burner centerline. For the lean equivalence ratios, this may be the result of the decrease

in flame speed as the equivalence ratio decreases. As the flame speed decreases the reaction zone will be anchored closer to the burner surface and more heat loss may occur.

CARS Measurements in the Room-Temperature Gas Cell

Dual-pump CARS measurements of the N₂/CO₂ spectra were acquired in a room-temperature gas cell at pressures up to 13 atm filled with a calibrated gas mixture. The gas mixture had a composition of 74% N₂ and 26% CO₂ on a mole fraction basis. CARS spectra acquired at 2.5 atm and at 12.7 atm are shown in Fig. 9. The theoretical fit to the experimental spectrum is shown along with the CO₂ mole fraction determined from the theoretical fit. The CO₂ mole fraction for the 2.5 atm spectrum shown in Fig. 6a is 0.29, reasonably close to the expected value of 0.26. As the pressure increases the accuracy of the CARS spectral model will become more questionable because of the severe collisional narrowing that occurs in all of the CO₂ bands. As can be seen from Fig. 9, the theoretical and experimental values of the relative intensities of the 00⁰0-10⁰0 and 00⁰0-02⁰0 CO₂ bands are in good agreement at 2.5 atm, but differ by approximately 50% at 12.7 atm. The best-fit value of the CO₂ concentration at 12.7 atm is 0.31, which is still in reasonable agreement with the actual value of 0.26.

CARS Measurements in the Swirl-Stabilized Combustor

A typical single-shot spectrum acquired in the exhaust region of a generic, atmospheric-pressure, swirl-stabilized combustor operating at an overall equivalence ratio of 0.41 is shown in Fig. 10. At the measured overall equivalence ratio of 0.41 for this test, the estimated value of the CO₂ mole fraction is 0.055. This

close agreement between the measured and predicted value of the CO₂ mole fraction is very encouraging. The temperature and CO₂ mole fraction pdfs resulting from the analysis of 500 single-shot CARS measurements are shown in Figs. 11 and 12, respectively. Of these 500 single-shot spectra, 355 were fit successfully using the CARS code; again the 145 spectra where CARSFIT returned the initial values of the fitting variables at the conclusion of the least-squares fitting process have not been examined in detail.

Measurements were also performed at overall equivalence ratios of 0.45, 0.50, 0.55, 0.60, 0.65, and 0.7. At equivalence ratios above 0.6 the heat load from the combustor misaligned the CARS optics; the combustor was shut down after the test at the 0.7 equivalence ratio and the CARS signal in room air had decreased by more than an order of magnitude. For future measurements in practical combustors it will be necessary to shield the CARS system from the combustor to maintain the system alignment.

For the measurements described in this paper, the CARS signals were obtained using the three-dimensional phase-matching scheme shown in Fig. 2. This phase-matching scheme gives in general the best spatial resolution of any CARS phase-matching scheme. However, the CARS signal level is more susceptible to beam steering caused by turbulence in the flow field when compared with other schemes such as annular phase matching. In annular phase-matching, which we have used in measurements in a high-pressure direct-injection natural gas engine (Green et al., 1998), the pump beams are propagated to the probe volume as an annular ring around the inner Stokes beam. The spatial resolution of this phase-matching scheme is approximately a factor of two worse than for three-dimensional phase-matching, but the

CARS signal level is much less susceptible to beam steering. Consequently, annular phase-matching may be a better phase-matching scheme for routine measurements in test combustors.

CONCLUSIONS

Dual-pump CARS measurements of temperature and CO₂ concentration were performed in (1) laminar propane/air flames, (2) hydrogen/air flames seeded with CO₂, (3) a room-temperature, high-pressure gas cell, and (4) a swirl-stabilized combustor fueled with JP-8. The single-shot capability of the dual-pump CO₂/N₂ CARS system was demonstrated in both the laboratory flames and in the swirl-stabilized combustor. In the steady laboratory flames, the standard deviations of the temperature and CO₂ mole fraction pdfs determined from the single-shot measurements were approximately 2% and 10% of the mean values, respectively. The spectral model for the CO₂ molecule in the Sandia CARSFIT code gave concentrations accurate to within approximately 10-20% in atmospheric pressure flames and in the room-temperature, high-pressure gas cell.

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REFERENCES

- M. Aldén and S. Wallin (1985), "CARS Experiments in a Full-Scale (10 m x 10 m) Industrial Coal Furnace," *Appl. Opt.* **24**, 3434-3437.
- R. R. Antcliff and O. Jarrett, Jr. (1987), "Multispecies Coherent Anti-Stokes

- Raman Scattering Instrument for Turbulent Combustion," *Rev. Sci. Instrum.* **58**, 2075-2080.
- P.-E. Bengtsson, L. Martinsson, M. Alden, B. Johansson, B. Lassesson, K. Marforio, and G. Lundholm (1994), "Dual-Broadband Rotational CARS Measurements in an IC Engine," pp. 1735-1742, *Twenty-Fifth Symposium (International) on Combustion*, The Combustion Institute, Pittsburgh, Pennsylvania.
- A. C. Eckbreth (1980), "CARS Thermometry in Practical Combustors," *Combust. Flame* **39**, 133-147.
- A. C. Eckbreth, G. M. Dobbs, J. H. Stufflebeam, and P. A. Tellex (1984), "CARS Temperature and Species Measurements in Augmented Jet Engine Exhausts," *Appl. Opt.* **23**, 1328-1339.
- A. C. Eckbreth, T. J. Anderson, and G. M. Dobbs (1988), "Multi-Color CARS for Hydrogen-Fueled Scramjet Applications," *Appl. Phys. B* **45**, 215-223.
- A. C. Eckbreth (1996), *Laser Diagnostics for Combustion Temperature and Species (Second Edition)*, Gordon and Breach Publishers, Amsterdam, The Netherlands.
- R. E. Foglesong, S. M. Green, R. P. Lucht, and J. C. Dutton (1998), "Dual-Pump Coherent Anti-Stokes Raman Scattering Technique for Simultaneous Measurement of Pressure and Temperature," *AIAA J.* **36**, 234-240.
- R. E. Foglesong (1999), private communication.
- S. Gordon and B. J. McBride (1976), *Computer Program for Calculation of Complex Chemical Equilibrium Compositions, Rocket Performance, Incident and Reflected Shocks, and Chapman-Jouget Detonations*, NASA Report SP-273.
- L. P. Goss, D. D. Trump, B. G. MacDonald, and G. L. Switzer (1983), "10-Hz Coherent Anti-Stokes Raman Spectroscopy Apparatus for Turbulent Combustion Studies," *Rev. Sci. Instrum.* **54**, 563-571.
- S. M. Green, P. J. Rubas, M. A. Paul, J. E. Peters, and R. P. Lucht (1998), "An Annular Phase-Matched Dual-Pump CARS System for the Simultaneous Detection of Nitrogen and Methane," *Appl. Opt.* **37**, 1690-1701.
- D. A. Greenhalgh, F. M. Porter, and W. A. England (1983), "The Application of Coherent Anti-Stokes Raman Scattering to Turbulent Combustion Thermometry," *Combust. Flame* **49**, 171-181.
- J. W. Hahn, C. W. Park, and S. N. Park (1997), "Broadband Coherent Anti-Stokes Raman Spectroscopy with a Modeless Dye Laser," *Appl. Opt.* **36**, 6722-6728.
- R. D. Hancock, F. R. Schauer, R. P. Lucht, and R. L. Farrow (1997a), "Dual-Pump Coherent Anti-Stokes Raman Scattering (CARS) Measurements of Hydrogen and Oxygen in a Laminar Jet Diffusion Flame," *Appl. Opt.* **36**, 3217-3226.
- R. D. Hancock, K. E. Bertagnolli, and R. P. Lucht (1997b), "Nitrogen and Hydrogen CARS Temperature Measurements in a Near-Adiabatic, Surface-Mixing (Hencken) Burner," *Combust. Flame* **109**, 323-331 (1997).
- R. P. Lucht (1987), "Three-Laser Coherent Anti-Stokes Raman Scattering Measurements of Two Species," *Opt. Lett.* **12**, 78-80.
- R. E. Palmer (1989), *The CARSFT Computer Code for Calculating Coherent Anti-Stokes Raman Spectra: User and Programmer Information*, Sandia National Laboratories Report SAND89-8206.

Frederick R. Schauer (1998), "Investigation of Flame Structure and Thermal Diffusion Effects in Hydrogen Jet Diffusion Flames," Ph.D. Thesis, University of Illinois at Urbana/Champaign.

G. Switzer, G. Sturgess, D. Sloan, and D. Shouse (1994), "Relation of CARS Temperature Fields to Lean Blowout Performance in an Aircraft Gas Turbine Generic Combustor," AIAA Paper No. AIAA 94-3271.

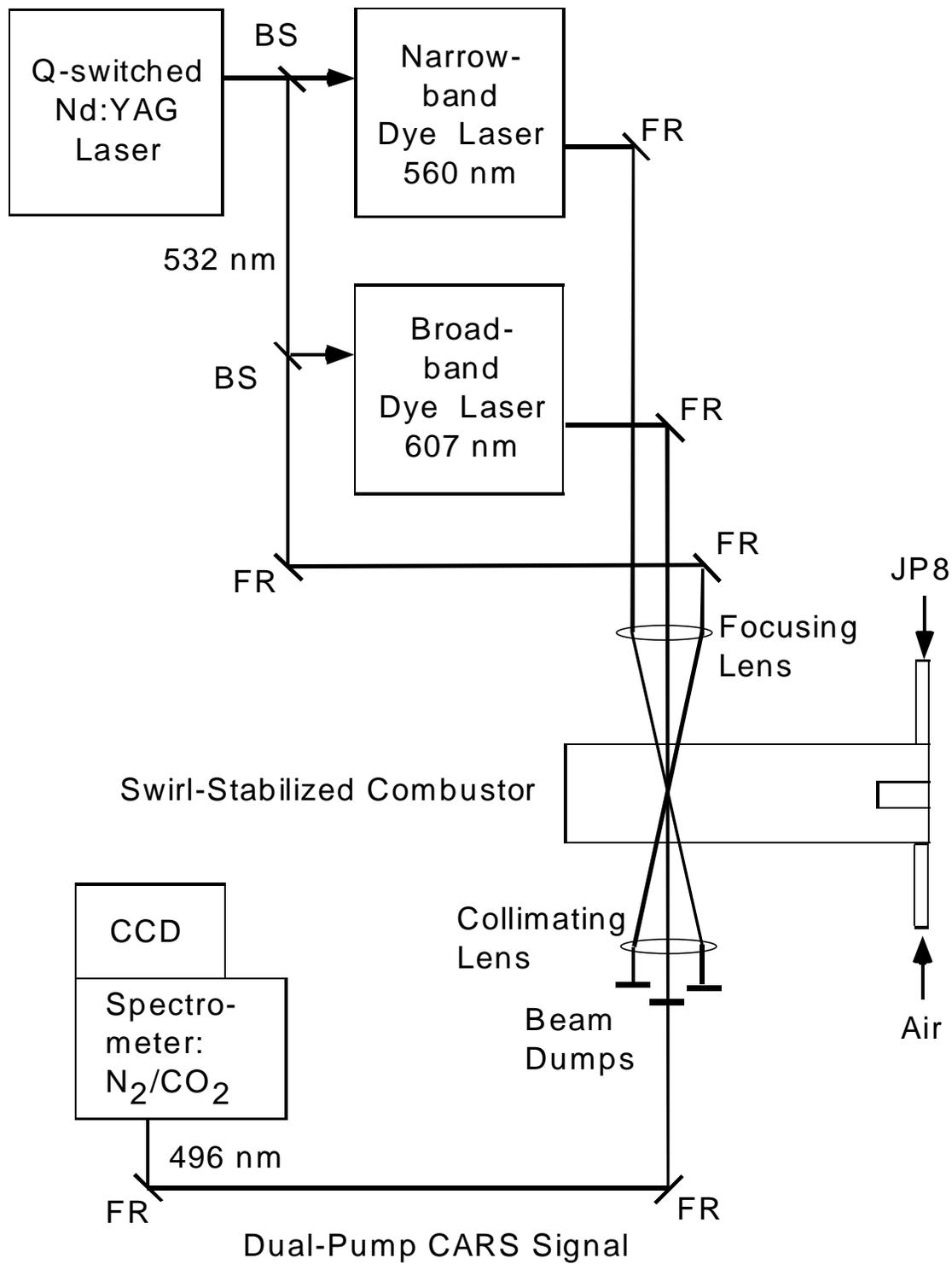


Figure 1. Experimental apparatus for the single-laser-shot, dual-pump N_2/CO_2 CARS system.

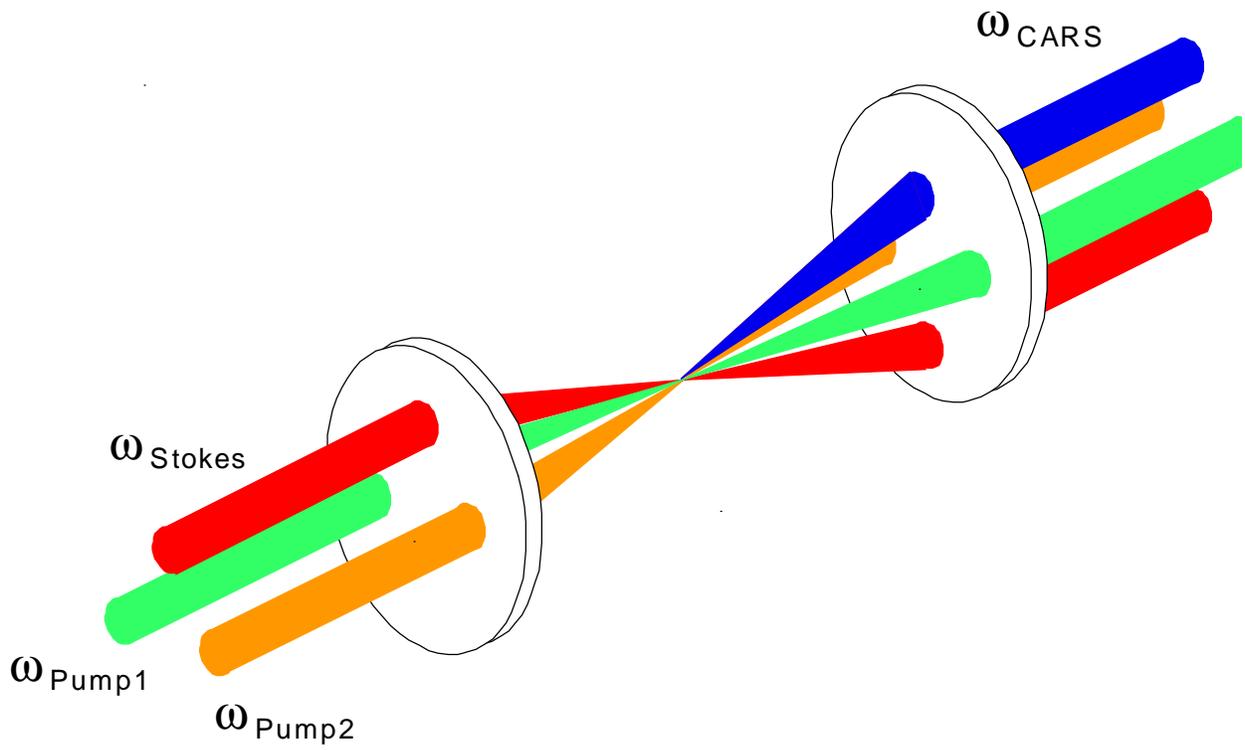


Figure 2. Three-dimensional phase-matching scheme used for the dual-pump N_2/CO_2 measurements.

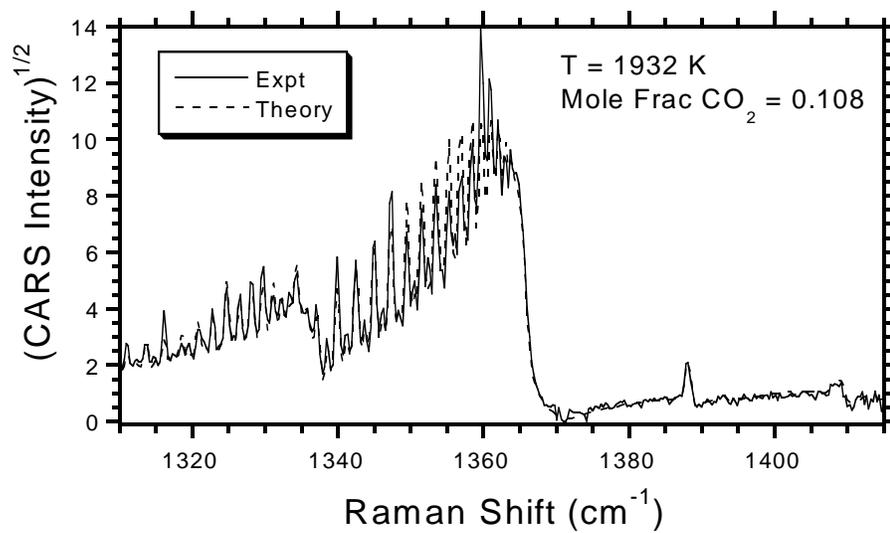
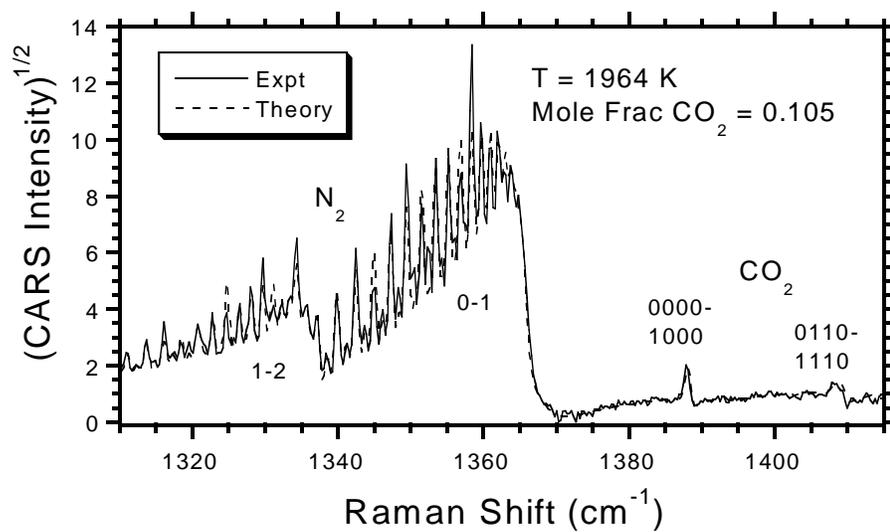


Figure 3. Single-laser-shot, dual-pump N₂/CO₂ spectra acquired from the propane/air flame.

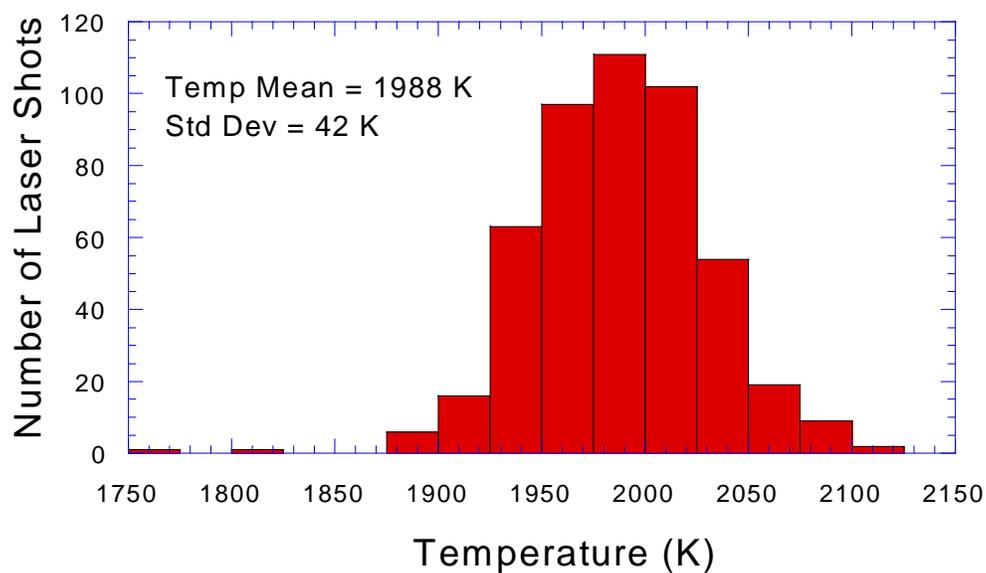


Figure 4. Probability distribution function of temperature determined from single-laser-shot, dual-pump N_2/CO_2 spectra acquired from the propane/air flame.

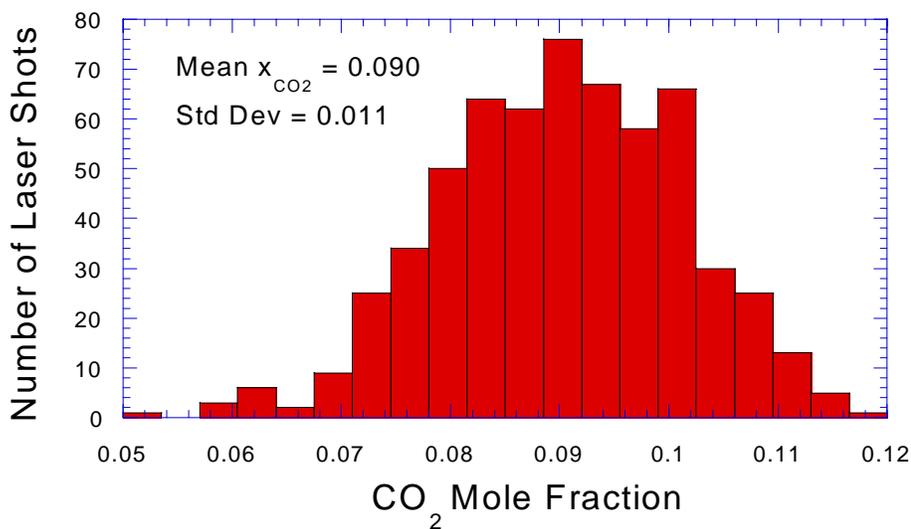


Figure 5. Probability distribution function of CO_2 mole fraction determined from single-laser-shot, dual-pump N_2/CO_2 spectra that were acquired in a stoichiometric propane/air flame.

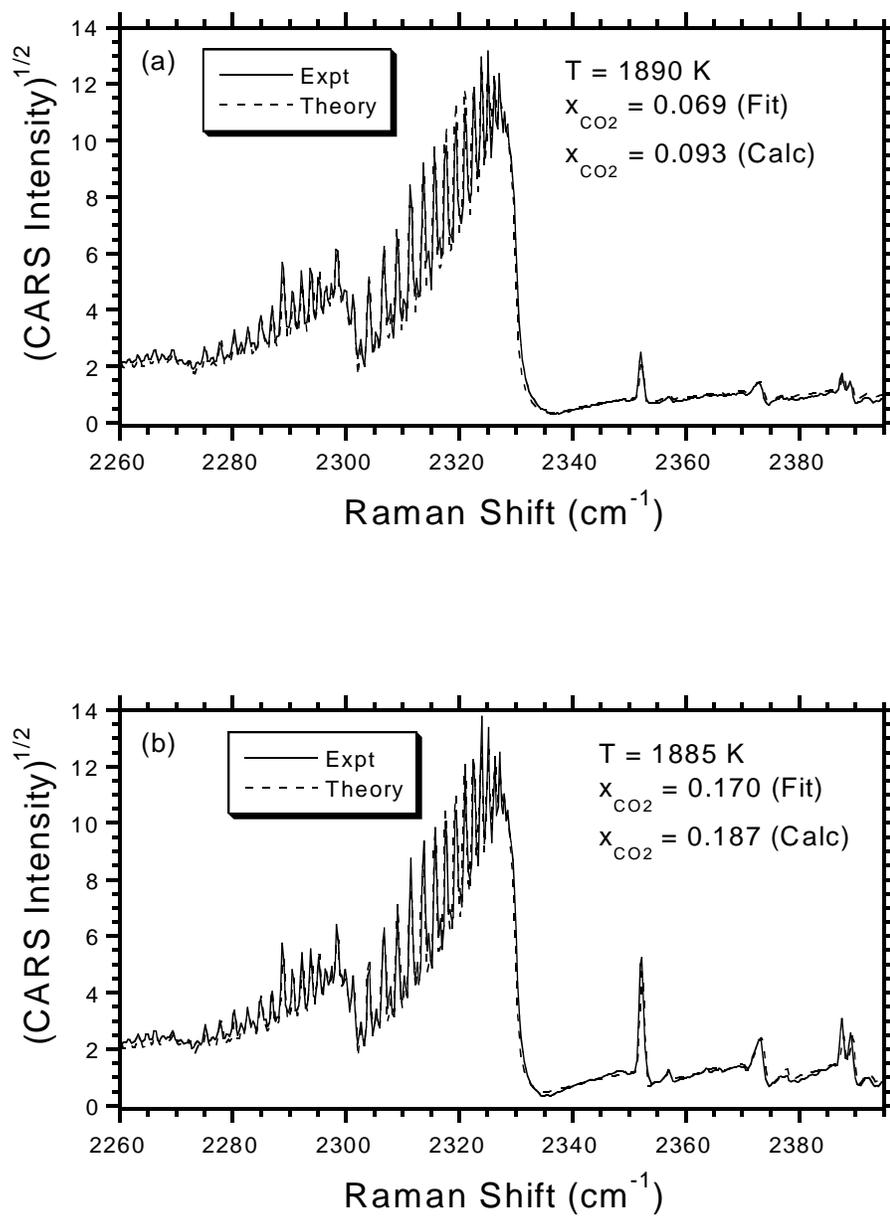


Figure 6. Dual-pump N_2/CO_2 spectra acquired in the propane/air/ CO_2 flames that were stabilized on the McKenna burner.

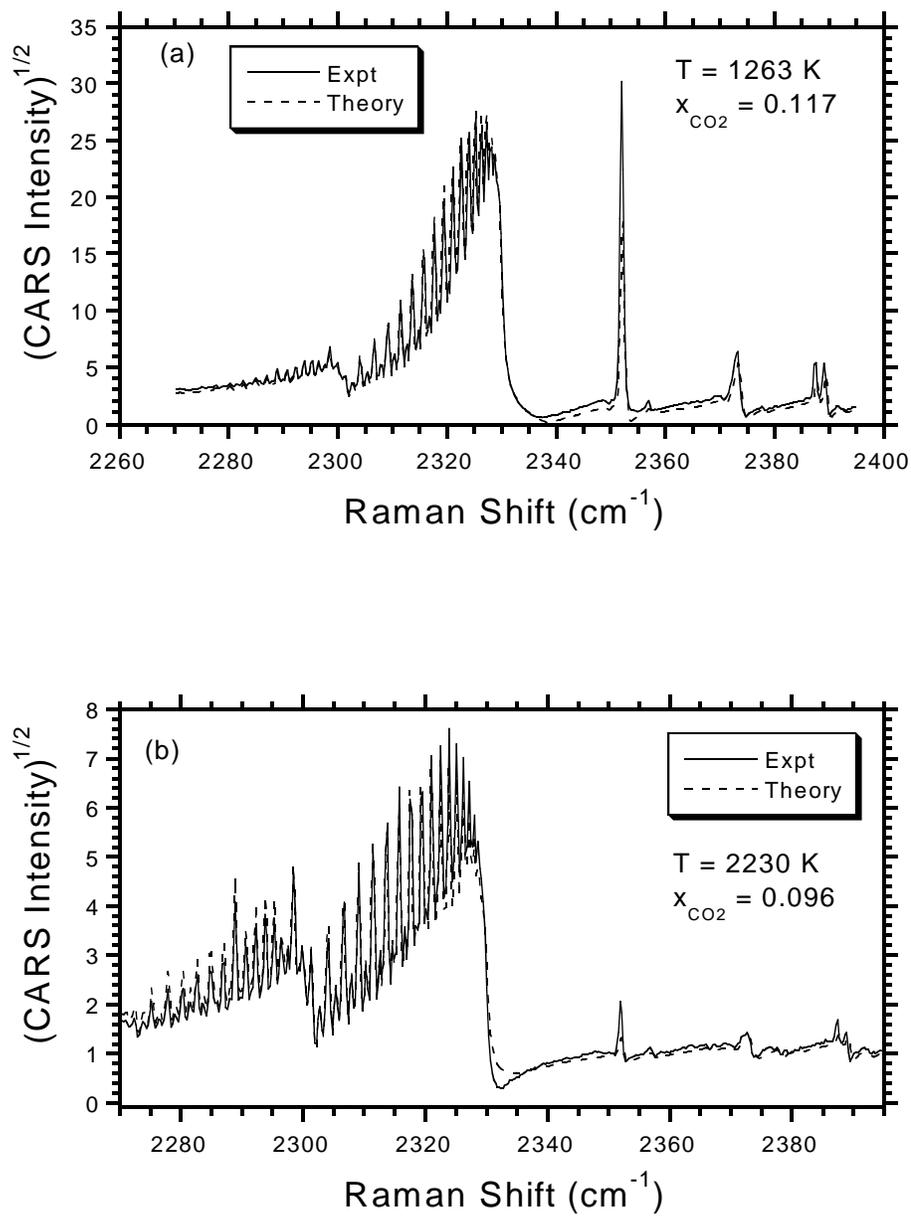


Figure 7. Dual-pump N_2/CO_2 spectra acquired in the near-adiabatic hydrogen/air/ CO_2 flames that were stabilized on the Hencken burner.

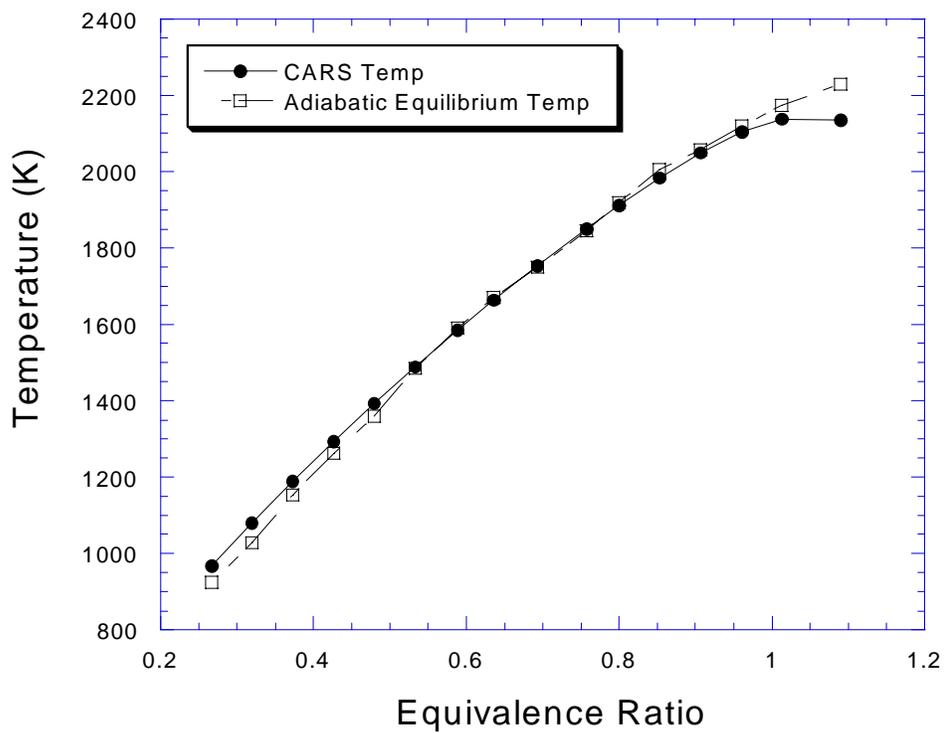


Figure 8. Comparison of measured CARS temperatures and calculated adiabatic equilibrium temperatures in the near-adiabatic hydrogen/air/CO₂ flames that were stabilized on the Hencken burner.

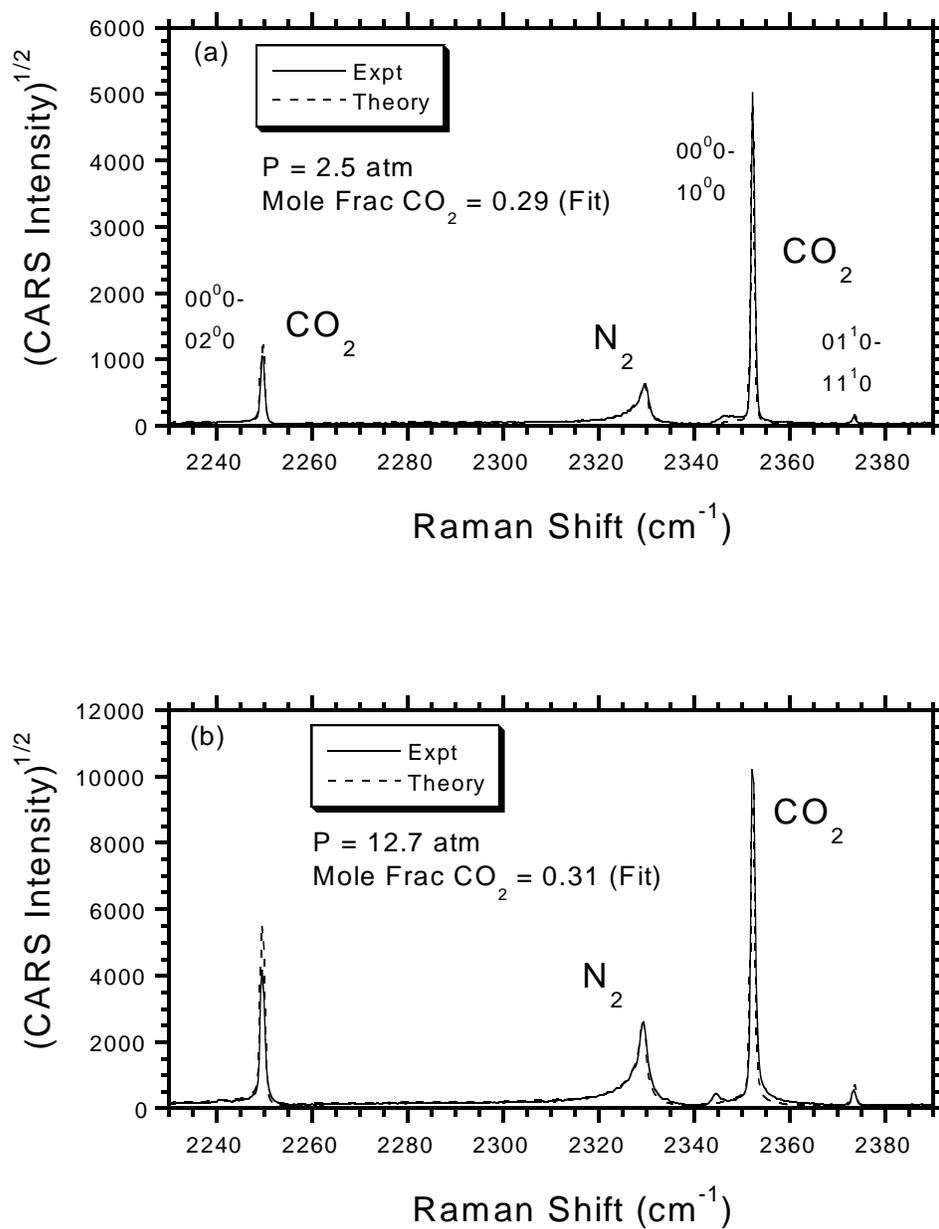


Figure 9. Dual-pump N_2/CO_2 spectra acquired in the room-temperature gas cell.

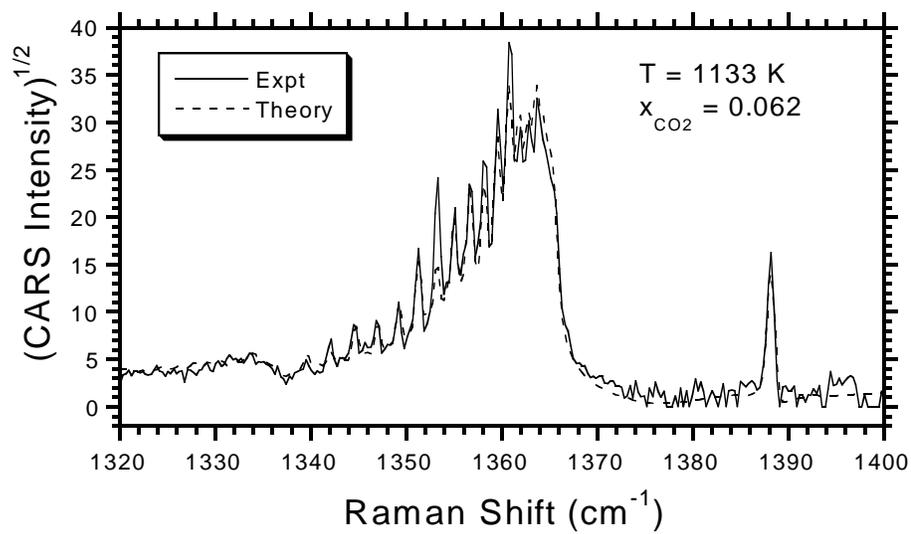


Figure 10. Single-laser-shot, dual-pump N_2/CO_2 spectra acquired in the exhaust region of the swirl-stabilized combustor.

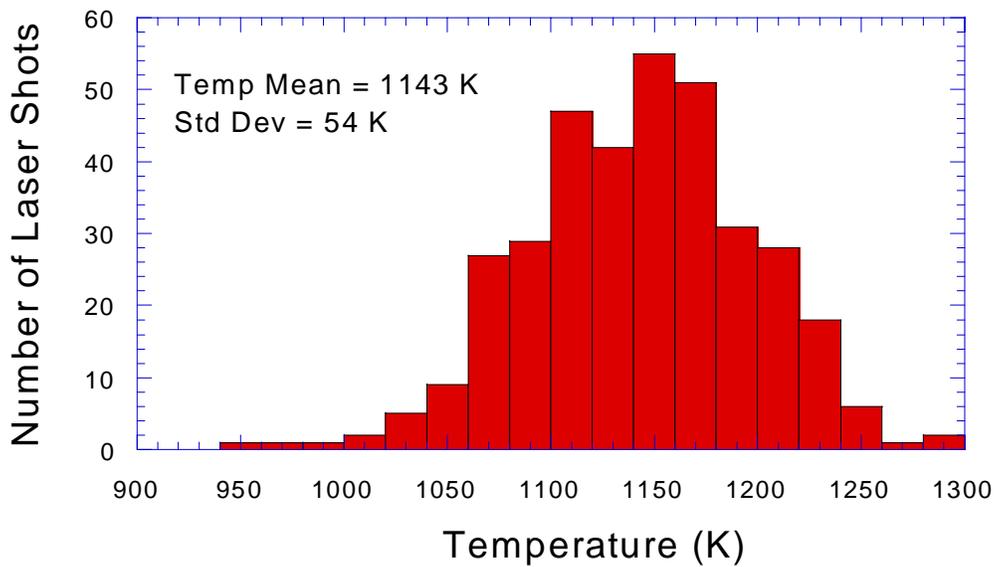


Figure 11. Probability distribution function of temperature determined from single-laser-shot, dual-pump N_2/CO_2 spectra that were acquired in the swirl-stabilized combustor.

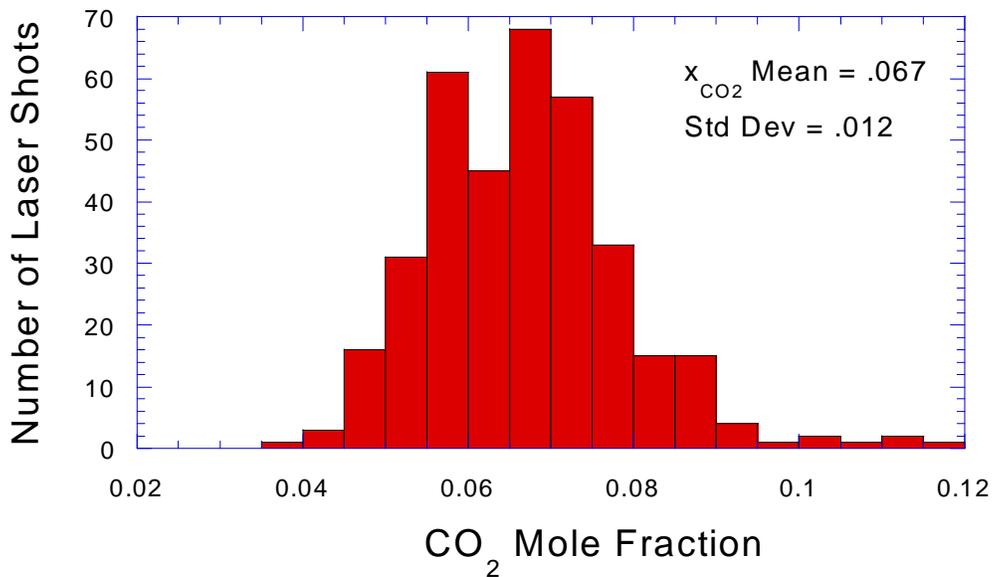


Figure 12. Probability distribution function of CO_2 mole fraction determined from single-laser-shot, dual-pump N_2/CO_2 spectra that were acquired in the swirl-stabilized combustor.