Dual-Pump Coherent Anti-Stokes Raman Scattering Temperature and CO$_2$ Concentration Measurements

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Measurements of temperature and CO$_2$ 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 U.S. Air Force Research Laboratory, Wright–Patterson Air Force Base. A modeless dye laser, a single-mode Nd:YAG laser, and an unintensified back-illuminated charge-coupled device digital camera were used for these measurements. The CARS measurements were performed on a single-laser-shot basis. The standard deviations of the temperatures and CO$_2$ mole fractions determined from single-shot dual-pump CARS spectra in steady laminar propane/air flames were approximately 2 and 10% of the mean values of approximately 2000 K and 0.10, respectively. The precision and accuracy of single-shot temperature measurements obtained from the nitrogen part of the dual-pump CARS system were investigated in detail in near-adiabatic hydrogen/air/CO$_2$ flames. The precision of the CARS temperature measurements was found to be comparable to the best results reported in the literature for conventional two-laser, single-pump CARS. The application of dual-pump CARS for single-shot measurements in a swirl-stabilized combustor fueled with JP-8 was also demonstrated.

Introduction

COHERENT anti-Stokes Raman scattering (CARS) has been applied widely for the measurement of temperature in a variety of practical combustors.[1-7] However, the use of CARS for measurements of species concentrations in combustor flowfields has been much less common.[8-13] Dual-pump CARS, demonstrated originally by Lucht,[14] is a technique whereby 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$_2$ (Refs. 14 and 15), N$_2$/H$_2$ (Ref. 16), N$_2$/CH$_4$ (Ref. 11), and N$_2$/CO$_2$ (Ref. 17) 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.[18]

Unlike other multispecies CARS techniques such as dual-Stokes and dual-broadband CARS,[1] 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 U.S. 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 are very useful for evaluating the combustion performance and efficiency of experimental combustors. The CARS spectra were analyzed using the Sandia National Laboratories CARSFIT code,[19] and experimental and theoretical CO$_2$ CARS spectra are compared. The temperature was determined from the N$_2$ spectrum because the CARS spectral model for N$_2$ has been much more thoroughly tested and validated than the spectral model for CO$_2$. In addition, the N$_2$ CARS signal is usually much stronger than the CO$_2$ CARS signal, and the rotational structure in the N$_2$ CARS spectrum can be resolved at atmospheric pressure. For these reasons, the N$_2$ spectrum also serves as a reference standard

against which to compare the measured and predicted intensities of the CO_2 spectral features.

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 Model GCR 5 injection-seeded, Q-switched Nd:YAG laser with a repetition rate of 10 Hz. The pulse energy of the 532-nm second-harmonic output of the Nd:YAG laser was approximately 600 mJ. A narrowband dye laser and a broadband dye laser were pumped using approximately 200 and 300 mJ, respectively, of the 532-nm radiation from the Nd:YAG laser. The narrowband dye laser was a Continuum Model ND6000, which has 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.[20] The spectral output of the broadband dye laser was centered at 607 nm, and the bandwidth was approximately 150 cm^{-1} full width at half-maximum.

The CARS signal was generated using the three dimensionally phase-matched arrangement shown in Fig. 2 (folded BOXCARS). The pulse energies for the 532-, 560-, and 607-nm beams at the CARS probe volume were typically 30, 30, and 15 mJ, respectively. The CARS focusing lens had a focal length of 400 mm. At the probe volume, the measured diameter of the 532-nm beam was approximately 90 ± 10 μm, whereas the 560- and 607-nm beam
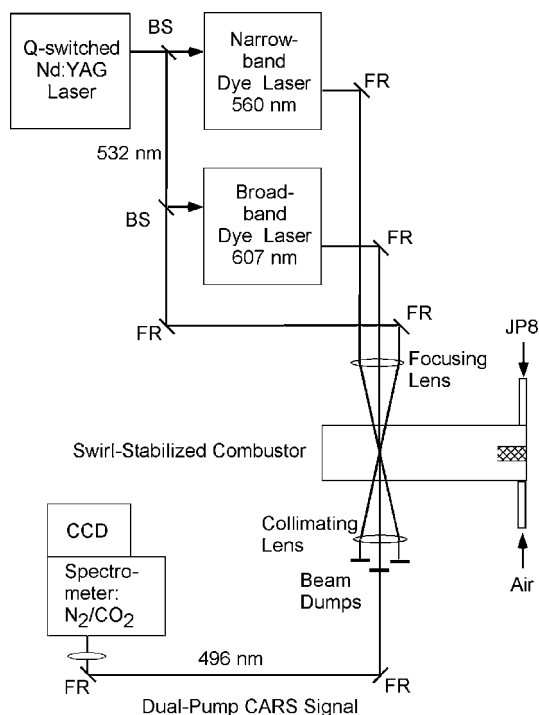
Fig. 1 Experimental apparatus for the single-laser-shot, dual-pump N_2/CO_2 CARS system.
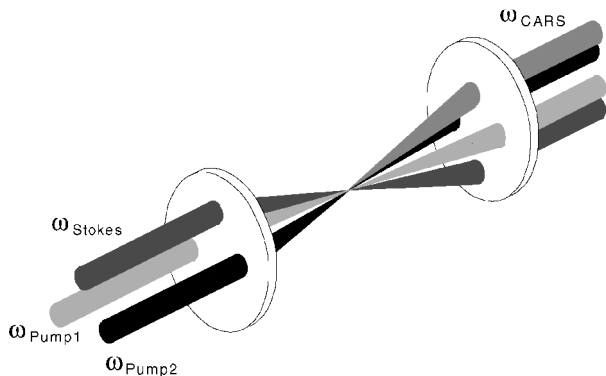
Fig. 2 Three-dimensional phase-matching scheme used for the dual-pump N_2/CO_2 measurements.

focal diameters were approximately 130 ± 15 μm. The spatial resolution of the CARS measurements is estimated to be approximately 2 mm, the interaction length over which approximately 80% of the CARS signal is generated. After passing through the CARS probe volume, the pump, Stokes, and CARS signal beams were recollimated using a 400-mm-focal-length lens. The pump and Stokes beams were directed into beam dumps, and the CARS signal beam was then focused onto the entrance slit of a SPEX 1-m spectrometer equipped with a 2400-line/mm holographic grating. The wavelength of the N_2 and CO_2 CARS signals was approximately 496 nm. The wavelength of the N_2 CARS signal was adjusted by tuning the wavelength of the narrowband dye laser. However, the wavelength of the CO_2 CARS signal did not vary as the narrowband dye laser wavelength was tuned. Therefore, the frequency separation of the N_2 and CO_2 CARS signals could be adjusted and optimized so that the main features for the two molecules did not overlap.

The CARS signal was detected using a 16-bit back-illuminated PixelVision SpectraVideo charge-coupled device (CCD) camera with a 165×1100 array of pixels (each pixel 24-μm 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 rate of 50,000 pixel/s. Even at this low readout rate, this camera is capable of achieving framing rates of $\sim$38 s^{-1} (reading out all 1100 elements), aided by a modification of the parallel-shift drivers to accommodate a 24-μs shift time. This thermoelectrically cooled CCD camera exhibited very low read noise (approximately 1.5 counts out of 65,536) and dark current (approximately 2 counts/s), whereas exhibiting a quantum efficiency of approximately 80% at 500 nm. Peak nitrogen CARS signals from the McKenna-burner flame ($T \approx 1900$ K) were typically 5000 counts, corresponding to approximately 40,000 photoelectrons. The Uniblitz 35-mm-aperture camera shutter was left open during data collection because it cannot operate at 10 Hz. Nonetheless, background flame emission, even in the case of the model gas-turbine combustor, did not warrant additional shuttering, apart from the readout process.

The camera and controller were linked to a personal computer with a PixelVision Lynx PCI serial card, which provides a fiber-optic link between the camera and the personal computer, a significant advantage for operation in large-scale facilities. The PixelView software package was employed to control the camera and collect CARS spectra. For each measurement (typically composed of 500–1000 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. Each spectrum was corrected afterward 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 both laminar propane/air flat-flame burners and a gas cell containing CO_2/N_2 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 CO_2, 3) the gas cell, and 4) the swirl-stabilized combustor was acquired. The CARS code was modified to allow batch processing of single-laser-shot spectra, greatly decreasing the time required to process the data.

The laminar flame and gas cell measurements were performed also 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 comparison of the experimental data acquired in these experiments with calculated results from the Sandia National Laboratories CARS code is of great interest.

CARS Spectral Model

The approach used to simulate the CO_2 CARS spectrum closely follows that of Hall and Stufflebeam.[21] Briefly, the 1200–1500 cm^{-1} region of the Raman spectrum of CO_2 is dominated by vibrational

transitions of the v_1 (symmetric stretch) and $2v_2$ (bending overtone) modes. Because of the near degeneracy $v_1 \cong 2v_2$, Fermi resonance couples certain sets of vibrational levels with quantum numbers differing by $\Delta v_1 = +1$ and $\Delta v_2 = -2$ (Fermi polyads). This coupling mixes the polyad vibrational states and significantly perturbs the frequencies and amplitudes of transitions involving the polyads. To describe these effects, we initially calculate unperturbed CO_2 vibrational levels using Hamiltonian expressions, force constants, and rotational constants from Suzuki[22] and Chedin.[23] The perturbed-state amplitudes and frequencies are then obtained by diagonalizing matrices involving the interacting states, using Fermi coupling constants from the same references. (The latter are evaluated only for the rotational quantum number J of the most populous level.) When possible, experimental vibrational energies and rotational constants from Rothman and Young[24] and Rothman[25] are used in place of the computed values, which are less accurate. In evaluating expressions for the third-order susceptibility, up to 128 rotational levels are included in each vibrational level. A total of 182 vibrational levels are considered for inclusion, based on population; consequently, simulations of flame spectra can include ≈ 7000 rotation–vibration levels. Only isotropic contributions from Raman Q-branch transitions ($\Delta J = 0$) are considered, and the isotropic polarizability derivatives are computed from Raman cross sections reported by Penney et al.[26] The transition collisional linewidths are assumed to be equal for all vibrational levels and determined entirely by rotational inelasticity, with J-dependent values computed from polynomial fits of theoretical Raman line-broadening coefficients of Rosenmann et al.[27] at 300 K. A temperature dependence of $(300/T)^n$, $n = 0.75$, was assumed; Hall and Stufflebeam[21] found no significant sensitivity of the simulated spectra to values of n in the range 0.5–1.0.

Collisional narrowing effects are approximated using a computationally efficient model based on the rotational diffusion theory of Gordon[28] (also see Ref. 29). Whereas more sophisticated collisional narrowing models based on full G-matrix approaches[30] have been developed for CO_2 CARS or Raman spectra, such models have been applied to individual bands of vibrational or bending transitions.[31,32] Because the broadband CARS spectral simulations necessary for this study typically involved tens to hundreds of vibrational bands, we did not employ the computationally intensive G-matrix method, which could potentially require separate matrix inversions and diagonalizations for each band. Finally, we used the spectral convolution expression given in Ref. 15 to account for instrument broadening contributed by the profiles of the narrowband dye laser and the spectrograph/array detector. (The linewidth of the injection-seeded Nd:YAG laser was much smaller than the Raman linewidths and was neglected.) The CARS spectral simulation for N_2 is based on susceptibility expressions and molecular constants described by Farrow et al.[33]

Experimental Results

CARS Measurements in Laboratory Flames

Some typical single-laser-shot spectra that were acquired in a stoichiometric propane/air flame stabilized on a water-cooled, sintered-bronze McKenna burner are shown in Fig. 3 along with the associated theoretical fits from the Sandia National Laboratories CARSFIT code. The wavelength of the narrowband dye laser that provided the second pump beam was adjusted to 561.0 nm to position the most prominent feature of the CO_2 Raman spectrum, the 00^00–10^00 band, close to the nitrogen bandhead. The abscissa in Fig. 3 corresponds to the Raman shift region for the CO_2 molecule. In Fig. 3, the Raman shift N_2 spectrum has been decreased by 964 cm^{-1}. In fact, the frequencies of the anti-Stokes signals in Fig. 3 range from 20,107 to 20,212 cm^{-1}, but the spectra are plotted vs Raman shift as is usual with CARS spectra.

As shown in Fig. 3, the theoretical fits to the single-shot spectra are excellent. For the stoichiometric propane/air flames investigated, the temperature is expected to be approximately 2000 K, somewhat below the adiabatic flame temperature due to heat loss to the burner. The equilibrium mole fraction of CO_2 is calculated to be 0.112 in the postflame region. Histograms of temperature and CO_2 mole fraction determined from 500 single-laser shots in the laminar propane/air flame are shown in Figs. 4 and 5, respectively.

a)

b)
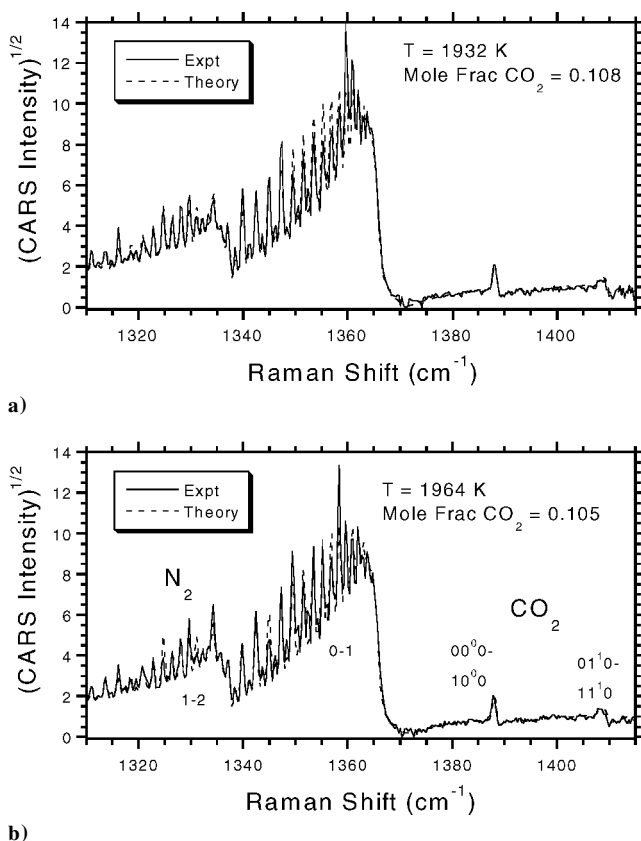
Fig. 3 Single-laser-shot, dual-pump N_2/CO_2 spectra acquired from a propane/air flame stabilized on a McKenna burner; temperature and CO_2 mole fractions determined from the CARS spectrum. Expected (adiabatic equilibrium) mole fraction is 0.103.
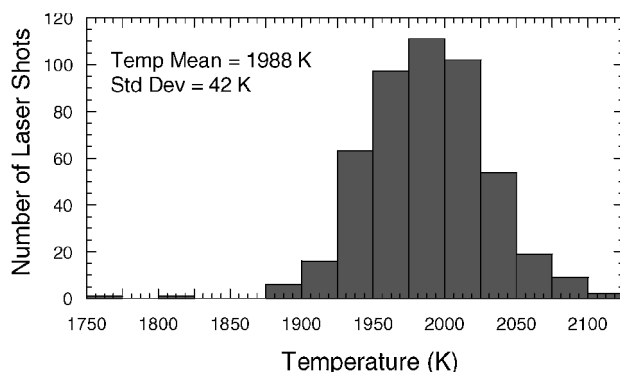

Fig. 4 PDF of temperature determined from single-laser-shot, dual-pump N_2/CO_2 spectra acquired from a stoichiometric propane/air flame.
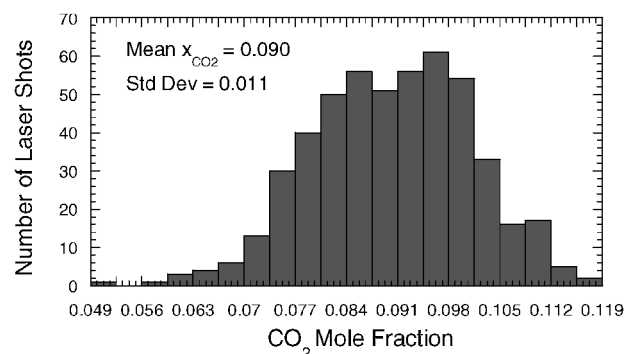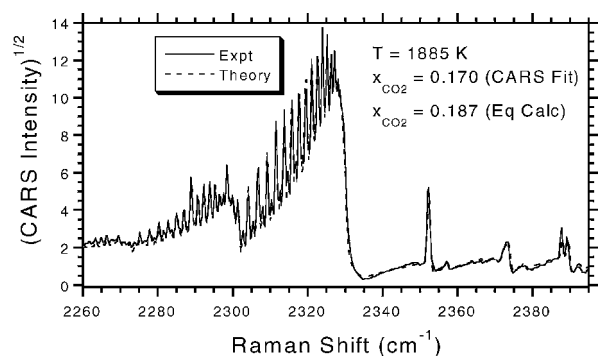

Fig. 5 PDF of CO_2 mole fraction determined from single-laser-shot, dual-pump N_2/CO_2 spectra acquired in a stoichiometric propane/air flame.

The probability density functions (PDFs) contain 480 points. 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 was not noticeably different. The mean and rms deviation of the temperature distribution are 1988 and 42 K, respectively. The mean and rms deviation of the CO_2 mole fraction distribution are 0.090 and 0.011, respectively. The measured mean CO_2 mole fraction is about 20% lower than the expected equilibrium value of 0.112, perhaps because some improvement is needed in the CO_2 CARS spectral simulation. Neglect of vibrational dephasing rate in the latter may account for much of the difference between the measured and predicted intensities of the CO_2 spectral features. The sensitivity of the CARS measurement of CO_2 is affected significantly because the rotational constants in the 00^00 and 10^00 vibrational levels are nearly equal. Consequently, the Q-branch rotational transitions are almost completely overlapped: The CO_2 CARS signal is enhanced greatly by this overlap and by significant collisional narrowing effects. Inadequacies of the Gordon diffusion model for collisional narrowing in CO_2 have been shown to underpredict theoretical peak intensities at elevated pressures and temperatures.[31] This tendency would lead to overprediction of best-fit CO_2 concentration if it occurred uniformly in the measured transitions.

Dual-pump CARS measurements were also performed in steady propane/air flames stabilized on a McKenna burner and doped with CO_2. The flow rates of propane and air were held constant, and the equivalence ratio was, therefore, constant; the flames were slightly fuel lean. The CO_2 flow rate was varied to obtain CARS spectra at different CO_2 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 CO_2 mole fraction is again somewhat lower than the equilibrium value based on the known flow rates, as was the case for the undoped flame described earlier. Note in Fig. 6 that the Raman shift of the CO_2 spectrum has been increased by 964 cm^{-1} and the N_2 and CO_2 spectra are plotted in the region of the N_2 Raman shift.
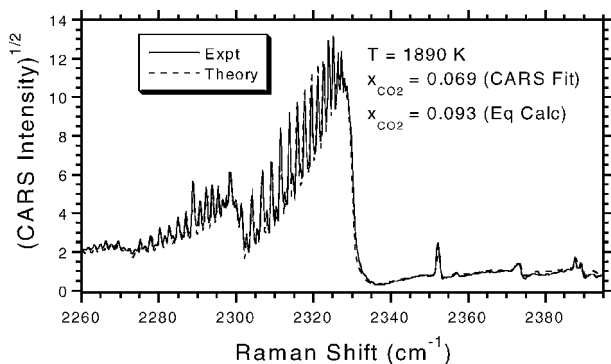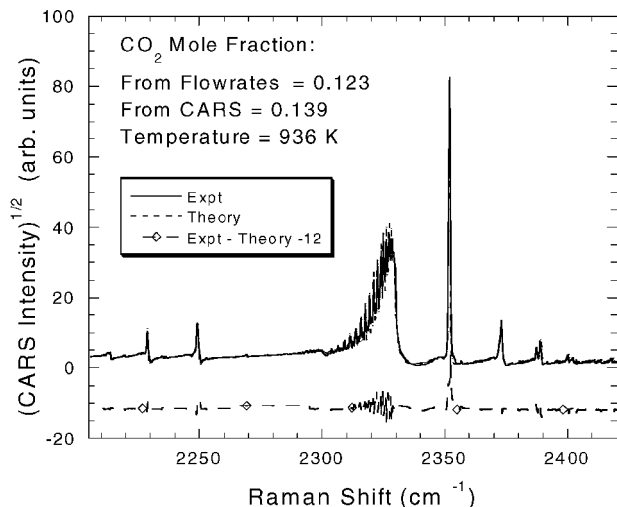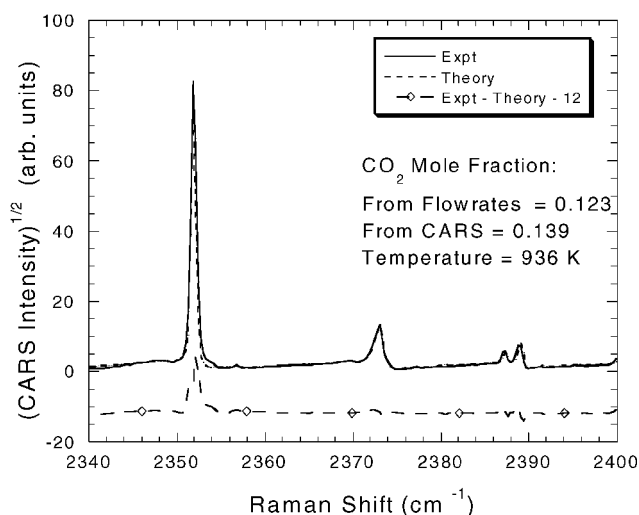

a)

b)

Fig. 6 Dual-pump N_2/CO_2 spectra acquired in propane/air/CO_2 flames stabilized on a McKenna burner, temperature and CO_2 mole fractions determined from the CARS spectrum, and expected equilibrium value for CO_2 mole fraction.


a) Overview

b) Expanded view

Fig. 7 Dual-pump N_2/CO_2 spectra acquired from near-adiabatic hydrogen/air/CO_2 flame stabilized on a Hencken burner at equivalence ratio of 0.26 with calculated adiabatic equilibrium temperature of 965 K.

Measurements were performed in near-adiabatic hydrogen/air/CO_2 flames stabilized on a Hencken burner to obtain dual-pump CO_2/N_2 CARS spectra over a wide range of temperatures.[34] The flow rate of the CO_2 was held constant while the hydrogen flow rate and, consequently, the temperature were 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 shown in Fig. 7, the spectra were acquired with very good signal-to-noise ratios. In our initial analysis of the Hencken burner spectra, we could not get good agreement between theory and experiment for the 00^00–10^00 band of CO_2. The reason for this difficulty is that we were trying to fit too wide a Raman shift range, and as a result, the spectral grid was too coarse for an accurate calculation of the intensity of the 00^00–10^00 band; this band is very narrow due to strong collisional narrowing effects. After this problem was realized, the central portion of the spectrum shown in Fig. 7a was fit for several different Raman shift ranges to ensure that the Sandia National Laboratories CARS code was calculating the band intensity in an accurate manner.

The temperatures determined from a least-squares fit of the nitrogen CARS spectrum and calculated using the NASA John H. Glenn Research Center at Lewis Field chemical equilibrium code[35] 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 ratios, this
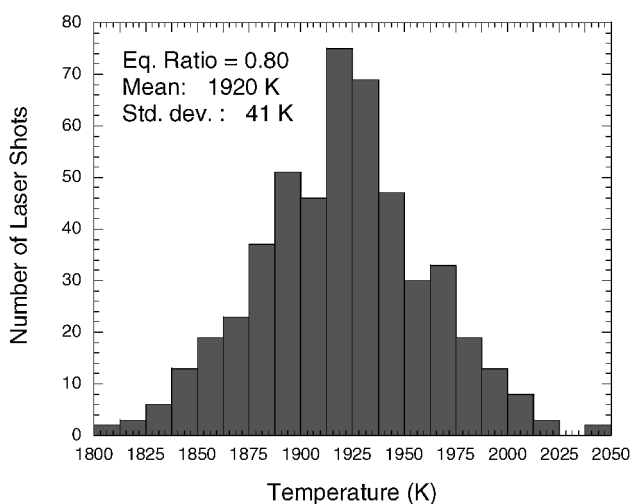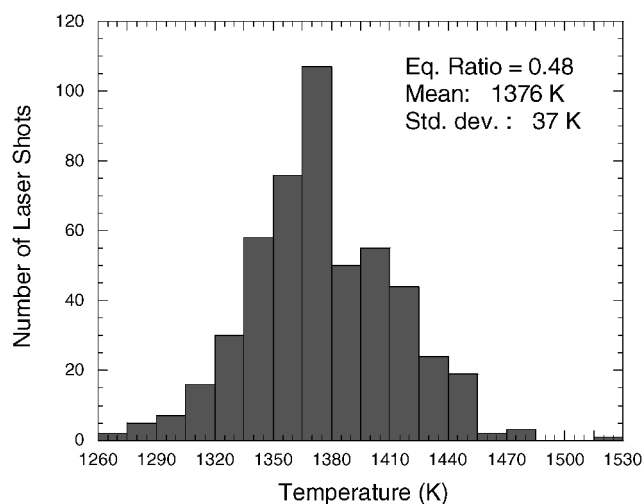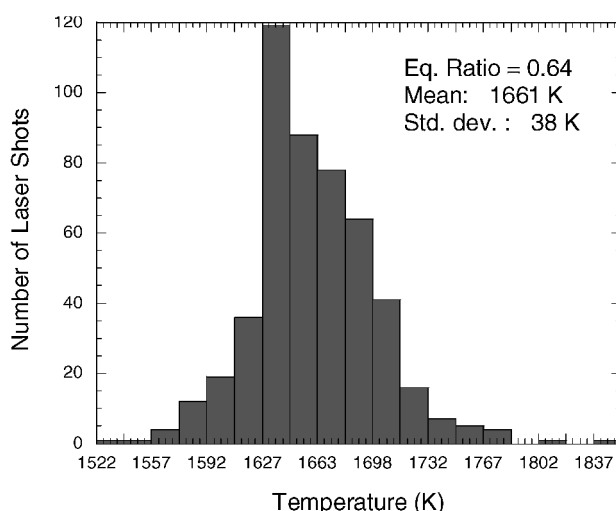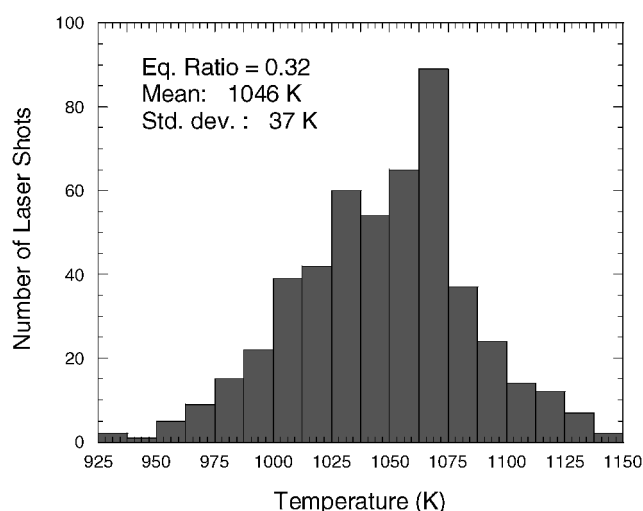
disagreement is probably the result of the slow rate of dissociation of the CO_2 molecule. For the flame with an equivalence ratio of 1.08, the calculated equilibrium mole fractions for CO_2 and CO are 0.0966 and 0.0176, respectively. The difference in the enthalpy of formation of the CO_2 and CO molecules could account for the superequilibrium temperatures that are observed, but full chemical kinetic modeling of the flames would be needed to verify this hypothesis.
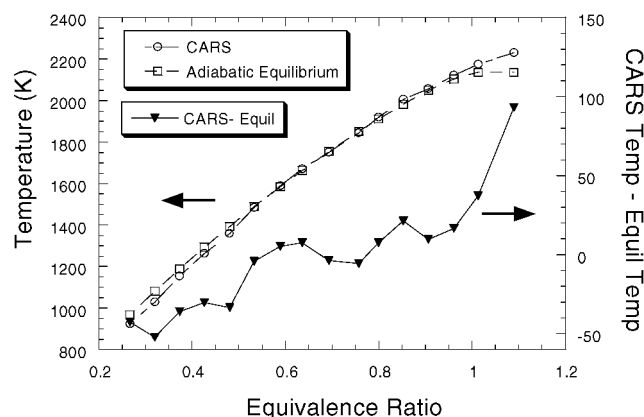
For the equivalence ratios less than 0.5, the measured temperatures are approximately 30 K less than the adiabatic equilibrium temperatures. This may be the result of the decrease in hydrogen jet velocity as the hydrogen flow rate is lowered to change the overall equivalence ratio. Flames are very stable on a Hencken burner because of the formation of diffusion flame zones at the exit of the hypodermic needles used to carry the fuel flow. As the fuel jet velocity decreases, there is likely to be more heat loss to the burner surface.

For each of the 16 equivalence ratios shown in Fig. 8, 1000 single-shot dual-pump CARS spectra were acquired. Temperature PDFs from four of these flames are shown in Fig. 9. For each condition, more than 990 of the 1000 single-shot spectra were fitted successfully using CARSFT. The mean and rms deviations for the temperature pdfs are 1046 ± 37, 1376 ± 37, 1661 ± 38, and 1920 ± 41 K for equivalence ratios of 0.32, 0.48, 0.64, and 0.80, respectively. The rms deviations for the temperature PDFs are comparable to the best results reported in the literature.[36−40] The temperatures were obtained by fitting only the nitrogen part of the spectrum. For the nitrogen CARS, the Raman polarization is created by the interaction of the single-mode 532-nm-pump beam and the modeless dye laser beam; the second, multimode pump beam from the narrowband dye laser is then scattered from the induced polarization to produce the CARS signal. It has been shown previously that for two-color CARS the rms deviation of the temperature PDF is reduced significantly by using a single-mode pump and modeless dye laser[36,37]; this also appears to be the case for our dual-pump CARS temperature measurements. The use of the back-illuminated unintensified CCD camera also eliminated the need for complicated calibration and correction procedures to account for the nonlinearity and limited dynamic

Fig. 8 Comparison of measured CARS temperatures and calculated adiabatic equilibrium temperatures in the near-adiabatic hydrogen/air/CO_2 flames stabilized on a Hencken burner.


Fig. 9   PDFs of temperature for four different flames determined from single-laser-shot, dual-pump N_2/CO_2 spectra acquired from the near-adiabatic hydrogen/air/CO_2 flames stabilized on a Hencken burner.

range of detectors employing microchannel plate intensifiers.[39] The effect of multimode as compared to single-mode pump radiation on the accuracy and precision of CARS temperature measurements is discussed in detail in Refs. 36–38 and 41–44.

CARS Measurements in the Room-Temperature Gas Cell

Dual-pump CARS measurements of the N_2/CO_2 spectra were acquired in a room-temperature gas cell at pressures up to 13 atm filled with a calibrated gas mixture with a composition of 74% N_2 and 26% CO_2 by volume. CARS spectra acquired at 2.5 and at 12.7 atm are shown in Fig. 10. The theoretical fit to the experimental spectrum is shown along with the CO_2 mole fraction determined from the theoretical fit. The CO_2 mole fraction for the 2.5 atm spectrum (Fig. 10a) is 0.29, reasonably close to the actual value of 0.26. As the pressure increases, the accuracy of the CARS spectral model will become more questionable because of the strong collisional narrowing that occurs in all of the CO_2 bands, which is only approximated by the Gordon[28] diffusion model. The neglect of vibrational dephasing is also a significant uncertainty in the CARS spectral model. More spectroscopic investigations are needed to refine the model in these respects, particularly for predicting relative intensities of different vibrational lines. As can be seen from Fig. 10, the theoretical and experimental values of the relative intensities of the 00^00–10^00 and 00^00–02^00 CO_2 bands are in good agreement at 2.5 atm, but differ by approximately 50% at 12.7 atm. Nonetheless, the best-fit value of the CO_2 mole fraction at 12.7 atm is 0.31, in reasonable agreement with the actual value of 0.26.
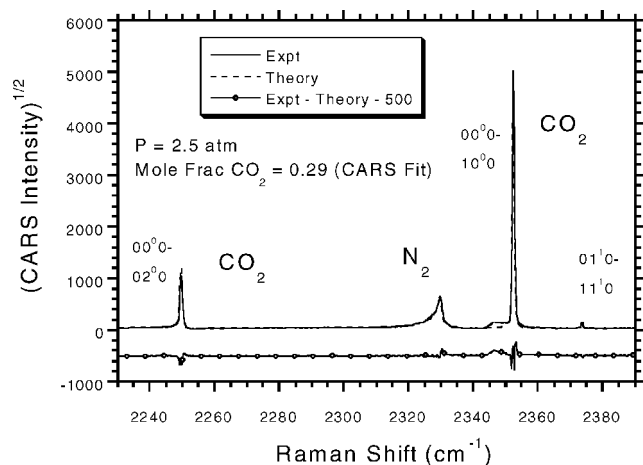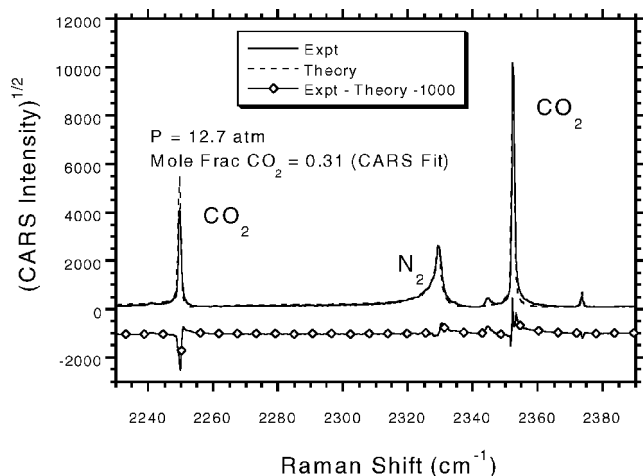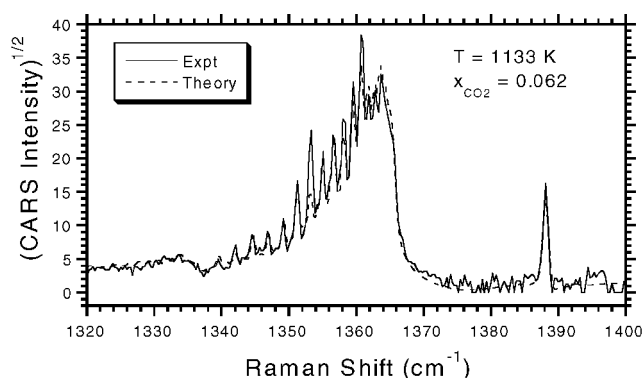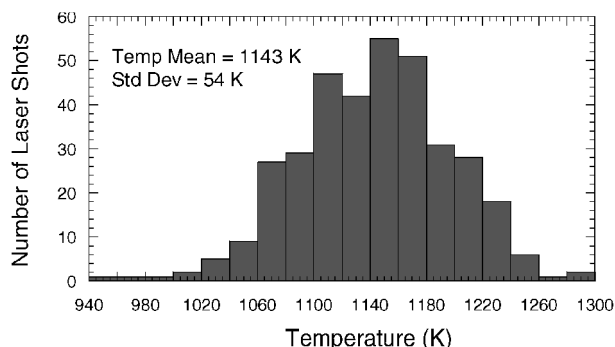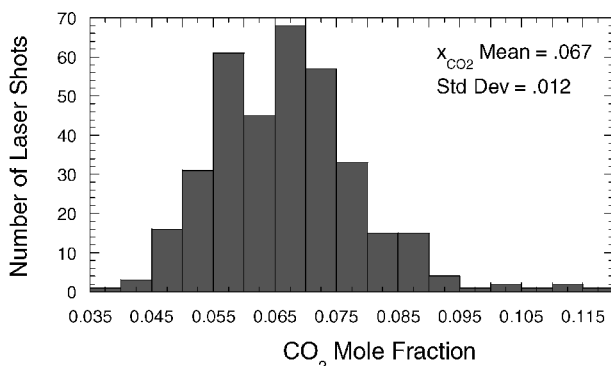

Fig. 10   Dual-pump N_2/CO_2 spectra acquired in a room-temperature gas cell where CO_2 mole fraction in calibrated gas mixture in the cell was 0.26.

Fig. 11   Single-laser-shot, dual-pump N_2/CO_2 spectra acquired in the exhaust region of the swirl-stabilized combustor operated at 0.41 overall equivalence ratio; temperature and CO_2 mole fraction values determined from fitting of CARS theoretical spectrum to experimental spectrum.


Fig. 12   PDF of temperature determined from single-laser-shot, dual-pump N_2/CO_2 spectra were acquired in the swirl-stabilized combustor operated at 0.41 overall equivalence ratio.

Fig. 13   PDF 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 operated at 0.41 overall equivalence ratio.

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. 11. The temperature and CO_2 mole fraction PDFs resulting from the analysis of 500 single-shot CARS measurements are shown in Figs. 12 and 13, respectively. Of these 500 single-shot spectra, 355 were fitted 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. At the measured overall equivalence ratio of 0.41, the mean value of the CO_2 mole fraction determined from the single-shot measurements was 0.067 with a standard deviation of 0.012. The mean value of the temperature determined from these same spectra was 1143 K with a standard deviation of 54 K. For combustion of JP-8 and air with an overall equivalence ratio of

a)

b)
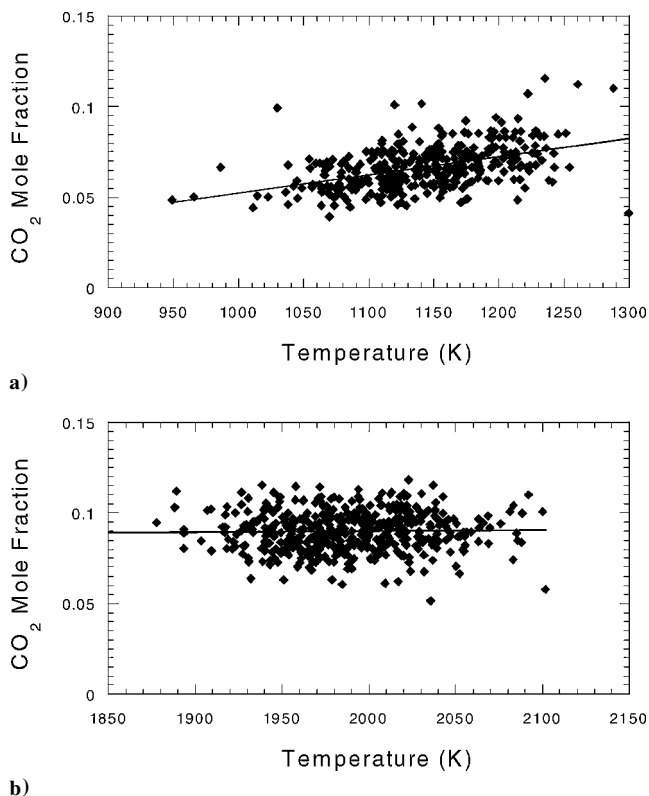
Fig. 14 Temperature–CO_2 mole fraction correlations for the single-shot CARS data a) acquired in the swirl-stabilized combustor operating at 0.41 equivalence ratio and b) in stoichiometric propane/air flame stabilized on a McKenna burner.

0.41, the adiabatic equilibrium values of CO_2 mole fraction and temperature are 0.057 and 1325 K, respectively. [The molecular formula of JP-8 is $C_{10.9}H_{20.9}$ and heat of formation is equal to -2.48×10^5 kJ/k mol (private communication, T. Edwards, U.S. Air Force Research Laboratory, Wright–Patterson Air Force Base, 2001).] The measured mean CO_2 concentration is approximately 20% higher than the adiabatic equilibrium value. This is the same level of agreement that was typical of the laminar flame studies. The 20% difference in the swirl-stabilized combustor studies may be due to inaccuracies in the measured fuel flow and airflow rates, to uncertainties in the CARS spectral model, or perhaps to averaging over inhomogeneities in the CARS probe volume. The measured mean temperature was almost 200 K lower than the adiabatic equilibrium temperature, presumably because of heat losses.

The correlation diagram for the measured CO_2 mole fraction and gas-phase temperature is shown in Fig. 14a. A least-squares fit line through the data shows a clear upward trend as expected; the higher CO_2 concentrations in the CARS probe volume indicate higher equivalence ratios, and the gas temperature is correspondingly higher. A similar correlation diagram for the stoichiometric propane/air laminar flame data is shown in Fig. 14b, and the least-squares-fit line through the data shows no such upward trend. The horizontal least-squares-fit line through the data in Fig. 14b indicates, as expected, that the laminar flame is steady and that the fluctuations in temperature and CO_2 mole fraction are due to the CARS instrumental apparatus.

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 CARS optics became misaligned significantly during the experiment because of the heat load from the combustor. For example, 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.

The CARS signals from the swirl-stabilized combustor 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 flowfield 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,[11] 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 somewhat 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

In the dual-pump CARS technique, signals from the two species of interest are generated by the same three laser beams. This allows the CARS spectra to be located at nearly the same frequency, simplifying their measurement, and provides for a high degree of correlation between the two signals, ensuring good single-shot fidelity for concentration measurements. Dual-pump CARS measurements of temperature and CO_2 concentration, employing a modeless dye laser, an injection-seeded Nd:YAG laser, and an unintensified back-illuminated CCD digital camera, were performed in 1) laminar propane/air flames stabilized on a water-cooled McKenna burner, 2) hydrogen/air flames seeded with CO_2 and stabilized on a near-adiabatic Hencken burner, 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 CO_2/N_2 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 CO_2 mole fraction PDFs determined from the single-shot measurements were approximately 2 and 10% of the mean values, respectively. Accuracy of the CO_2 mole fractions derived from the CARS signals and the Sandia National Laboratories CARS code was reasonably good, about 10–20% in atmospheric-pressure flames and in the room-temperature, high-pressure gas cell, in spite of strong collisional narrowing of the CO_2 lines. The limited parameter range that was investigated in these measurements indicates the need to investigate the CARS spectrum of CO_2 over much wider ranges of pressure and temperature. The dual-pump CARS technique is ideal for these types of investigations because the nitrogen CARS spectrum can be used as a concentration reference for the CO_2 spectrum, for thermometry, and for accurate characterization of experimental parameters such as the instrument function for the spectrometer–CCD system.

These measurements demonstrate also the potential of multipump CARS techniques for the characterization of the temperature and the major species concentrations in challenging environments such as model gas-turbine combustors. The high degree of correlation between the nitrogen signal and the CO_2 signal in the dual-pump CARS technique is an especially valuable characteristic of the measurements. Strong collisional narrowing in the 00^00–10^00 band increases the intensity of this band significantly even at atmospheric pressure, allowing the measurement of single-shot CO_2 CARS signals at mole fractions down to the level of 1–2%. The dual-pump N_2/CO_2 CARS system will be useful for investigating a wide variety of hydrocarbon-fueled combustion systems.

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