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DUAL-PUMP COHERENT ANTI-STOKES RAMAN SCATTERING THERMOMETRY IN A SOOTING TURBULENT POOL FIRE

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Abstract

We present a dual-pump coherent anti-Stokes Raman scattering (CARS) instrument, which has been constructed for the probing of temperature fluctuations in turbulent pool fires of meter-scale. The measurements are performed at the newly commissioned Fire Laboratory for Accreditation of Models and Experiments (FLAME) facility at Sandia National Laboratories, which provides a canonical fire plume in quiescent wind conditions, with well-characterized boundary conditions and access for modern laser-diagnostic probes. The details of the dual-pump CARS experimental facility for the fire-science application are presented, and single-laser-shot CARS spectra containing information from in-fire N_2, O_2, H_2, and CO_2 are provided. Single-shot temperatures are obtained from spectral fitting of the Raman *Q*-branch signature of N_2, from which histograms that estimate the pdf of the temperature fluctuations at the center of the fire plume are presented. Results from two different sooting fire experiments reveal excellent test-to-test repeatability of the fire plume provided by FLAME, as well as the CARS-measured temperatures. The accuracy and precision of the CARS temperatures is assessed from measurements in furnace-heated air, where the temperature can be accurately determined by a thermocouple. At temperatures in excess of 500 K, the furnace results show that the CARS measurements are accurate to within 2-3% and precise to within ±3-5% of the measured absolute temperature.

Keywords: Fire Science, Thermometry, Laser Diagnostics, CARS

1. Introduction

Fire is the dominant source risk to personal, commercial and military assets, and effective risk-management approaches will increasingly rely upon an improved understanding of fire environments and predictive fire-simulation tools. The growing need to understand the fundamental nature of fire has lead to recent advancements in optical-diagnostic tools for fire applications as replacements to traditional, but limited, physical probes. For fire testing, optical methods offer the potential for simultaneous measurement of multiple thermochemical parameters with minimal sensor intrusion and very fine spatio-temporal resolution. Many of the laser-diagnostic techniques that have revolutionized laboratory combustion research [1, 2] have matured, and some approaches have been shown to offer additional advantages for fundamental studies of large-scale combustion systems. Examples specific to fire research include particle-image velocimetry and planar laser-induced fluorescence for 2-D velocity and scalar measurements in meter-scale buoyant plumes and gas-fueled fires [3, 4]; species detection with tunable-diode-laser absorption spectroscopy [5]; as well as thermometry using IR emission spectroscopy [6]; and *in situ* pyrometric probes [5, 7].

Coherent anti-Stokes Raman scattering (CARS) is a laser diagnostic with great potential for large-scale fire testing and research. The CARS technique has a long history of robust application in large-scale practical combustion systems, and has been used since the 1980's in field applications, including jet engine exhausts [8], coal-fired furnaces [9], and full-scale MHD combustors [10]. Vis-à-vis the approaches cited above, CARS is capable of providing temperature and species measurements with improved spatial resolution, which are free of the inherent uncertainties in soot and species radiative and optical properties. The strength of CARS signatures permits single-laser-shot measurements for characterization of highly fluctuating turbulent fire environments, and CARS is one of only a small handful of optical diagnostic techniques capable of providing spatially and temporally resolved temperature and species measurements in the heavily sooting flames which are characteristic of many hydrocarbon fire systems [11-13]. We have recently reported [14] what we believe to be the first application of CARS to fire testing. This early work implemented a conventional approach to CARS of the N_2 molecule, in which frequency degenerate pump beams from the Nd:YAG laser were employed for thermometry in a non-sooting methanol-fueled pool fire. While effective, the degenerate-pump-beam approach is limited to probing of soot-free fire environments, as significant interference from Swan-band transitions in laser-produced C_2 [15] is spectrally coincident with the N_2 CARS signature near 473 nm. In this paper, we report a recent upgrade of our CARS capability for fire testing to the dual-pump CARS approach [16], and the application of our dual-pump instrument for thermometry in a sooting fire plume. The dual-pump approach allows the CARS signal to be shifted to an arbitrary spectral position that is free of interference from the C_2 radical [12, 13] and also permits simultaneous acquisition of spectra from O_2, H_2, and CO_2 species, from which mole-fraction data may be obtained simultaneously with the temperature.

2. FLAME Facility

Our measurements were performed at Sandia's Fire Laboratory for Accreditation of Models and Experiments (FLAME) facility. FLAME allows for liquid or gaseous fueled fires up to 3 m in base diameter to be studied in a well-controlled environment. The facility has been specifically designed to accommodate laser diagnostics, which provides a unique opportunity for field-scale fire testing with control of the experiment that is typically achieved only in the laboratory. A drawing of the FLAME test bay is provided in Fig. 1. The walls of the test bay are water cooled to provide a well-controlled, ambient-temperature radiative boundary condition, and the liquid- or gas-fueled burner sits in the center of the grated-steel floor of the cylindrical test bay that is 18.3 m in diameter and 12.2 m in height. The ceiling slopes upwards (~18º) from the perimeter walls to a height of 14.6 m over the center of the facility, where the air and combustion gases transition to a 3.0-m × 3.7-m square chimney that exhausts to an electrostatic precipitator where soot is removed before the gas stream exits through a smoke stack. Combustion air enters the facility basement level via a 3.05-m-diameter pipe and is distributed by 18 supply lines to an annular air ring along the circumference of the basement floor. Flow through the air ring was uniform within 10% of the mean flow. The fire draws in combustion air through the grated floor by natural draft, and the air flows horizontally along a 12-m-diameter steel skirt, which surrounds the fuel pan and forms the fire ground plane, before it is entrained into the fire plume.

A digital photograph of the sooting pool fire investigated is shown in Fig. 2. For the experiments reported here, a 2-m-diameter × 50-mm-deep steel fuel pan is mounted at the facility center, with the edge of the pan flush with the fire ground plane. The liquid fuel was a 90% methanol / 10% toluene by volume blend, which was premixed and stored in large drums. Only enough fuel to sustain 15-20 minutes of steady state burn was added, such that the liquid surface was never flush with the top of the fuel pan. No make-up fuel was added during the experiments, and the fires were typically run until all of the fuel was consumed. In the absence of a direct soot measurement, the soot volume fraction in this fire was estimated to be of order 10^{-7} using the known smoke yields of the pure fuels. The fire provided significant visible luminosity from soot, while remaining sufficiently transparent for propagation of the CARS signal and pump/Stokes laser beams. The laser beams and CARS signal were propagated through insulated steel “light pipes,” seen in Fig. 2, to limit the path length through the fire for beam steering and absorption of light by soot. These pipes were 101 mm in diameter and 67 cm long, leaving a 66-cm path through the fire for the laser beams to travel. The light pipes also served the additional purpose of shielding any high-energy laser optics that were placed in close proximity to the fire.

3. Dual-Pump CARS Technique and Optical System

The physics of the dual-pump CARS approach, as first demonstrated by Lucht [16], are summarized in the energy level diagrams shown in Fig. 3. Experimentally, three pulsed laser beams—termed "pump-1" , "pump-2", and "Stokes"—at frequencies ω_1, ω_2, and ω_S are focused and crossed at a high-intensity spot in the three-dimensional crossed-beam geometry [17] shown in Fig. 4. The center frequency of a broadband dye-laser Stokes source is tuned so that the frequency difference, $\omega_1 - \omega_s$, is coincident with the vibrational Raman frequencies of N_2; this tuning induces an oscillating polarization in the measurement volume that scatters the ω_2 photons to deliver a coherent, laser-like CARS signal beam at frequency $\omega_{CARS} = (\omega_1 - \omega_s) + \omega_2$, as illustrated in Fig. 4. For measurements in sooting flames, a tunable narrowband dye laser is used to scan the frequency, ω_2, which positions ω_{CARS} in a portion of the visible spectrum that is free of interference from laser-produced C_2 and other olefinic species [13]. In our measurements, a pump-2 wavelength of λ_2 = 561.14 nm was used, which positioned the bandhead of the N_2 CARS signal beam near 496.2 nm, and additionally tuned the $\omega_2 - \omega_S$ frequency difference to drive Raman polarizations in CO_2, H_2, and O_2 which scattered ω_1 photons to yield additional CARS signatures from these species which were in close spectral proximity to the N_2 CARS signature. With this approach, temperature is readily determined from the shape of the N_2 *Q*-branch signature and gas-phase species information from the relative heights of the N_2, CO_2, H_2, and O_2 peaks in the CARS spectrum.

A dual-pump CARS instrument has been fielded in one of the three optics laboratories adjacent to the FLAME test bay, as shown in Fig. 5. A frequency doubled, *Q*-switched Nd:YAG laser (Continuum PowerLite Plus) operating at 10 Hz supplies 1,700 mJ per 8-ns pulse at a wavelength of 532 nm. With injection seeding, the output of the Nd:YAG source has a nearly transform-limited linewidth of 0.003 cm^{-1}. The Nd:YAG system provides sufficient energy to pump both narrowband tunable dye laser (pump-2 source) and the broadband Stokes dye source, while also supplying the pump-1 beam to the

CARS process. The broadband Stokes laser output is optimized for probing of the N_2 molecule with the pump-1/Stokes laser pair by using a dye mixture of Rhodamine 610 and Rhodamine 640 in methanol to provide a Stokes output centered at 607 nm with a nominal bandwidth of 225 cm^{-1} (FWHM), as measured by fits to nonresonant CARS spectra obtained from argon. The narrowband dye source (Continuum ND6000) utilizes Rhodamine 590 dye in methanol to supply tunable pump-2 radiation with a with a 0.08 cm^{-1} manufacturer-specified linewidth at a wavelength of 561.14 nm. The energy in the Stokes and pump beams is controlled by combinations of half-wave rotators and polarizers throughout the optical system. Nominal beam energies are 50, 50, and 30 mJ/pulse for the pump-1, pump-2, and Stokes sources, respectively. These high pulse energies were required to obtain sufficient single-shot signal strength; measurement bias due to Stark effect and stimulated Raman pumping [18] at high pulse energies was checked by performing shot-averaged CARS measurements in a premixed CH_4/air flame stabilized on a near-adiabatic flat-flame burner [19], and systematically varying the pulse energies. This process revealed no systematic change in the CARS measured temperatures with increasing laser power.

The pump and Stokes beams are aligned in the folded BOXCARS configuration shown in Fig. 4 and the phase-matched beam configuration was delivered to the FLAME test bay using a series of mirrors and periscopes. Fine-scale alignment of all three laser beams is provided by mounting a single turning optic in each beam line with a high-precision tip-tilt mount (Newport # 610) atop a micrometer-driven linear translator. These mounts permit optimization of the CARS signal-to-noise (SNR) as they enable us to make μradian-scale adjustments to the beam paths while observing the growth of the CARS signal. Inclusion of these high-precision mounts into the instrument is crucial because the long path length (~11 m) between the CARS measurement volume and the west laboratory optical tables greatly amplifies any movements made in the laser laboratory.

The laser beams are mirror coupled to the test bay and propagate ~8.5 m across the facility to an *f* = 1000 mm singlet beam-crossing lens which focuses the pump and Stokes beams at a CARS measurement volume that is ~1 m above the liquid fuel surface and above the center of the fuel pan. The beam-crossing lens is housed in an insulated steel enclosure adjacent to a 10.1-cm-diameter × 67-cm-long steel light pipe through which the laser beams propagate as they focus. The CARS signal is generated within a ~100-200-μm diameter × 10-mm long measurement volume at the center of the fire plume. The CARS signal and pump/Stokes laser beams propagate through the second steel light pipe to an *f* = 1000 mm collimating lens, and the collimated signal beam is separated from the high-intensity laser beams by a series of six dichroic beam splitters and a hard-coated interference filter with > 90% transmission at the CARS signal wavelength of ~496 nm. The CARS beam is transmitted by a 30-m long fiber of 100-μm core which was directly coupled to the 50-μm entrance slit of a spectrograph located in the optical laboratory. Transmission of the CARS beam through the low-loss fiber is in excess of 80%, with losses primarily resulting from the coupling process. The 0.75-m long spectrograph with 1200 l/mm grating disperses the CARS signal onto a back-illuminated CCD camera with ~95% quantum efficiency at the CARS-signal wavelength. A magnification of 3.75 at the CCD focal plane is provided by a relay lens pair at the exit of the spectrograph, which results in a detection system dispersion of ~0.30 cm^{-1}/pixel and a nominal system resolution of 2.1 cm^{-1}, as measured from spectra obtained using a Xe calibration lamp. Overfilling of the *f*/7 spectrograph by the *f*/2.3 was minimized by coupling of the CARS signal to low-order fiber modes using slow, *f*/10 optics to launch the CARS signal into the fiber.

Ensembles of several thousand single-laser-shot CARS spectra were acquired at the single measurement point during the nominally 15-20 minute pool-fire burns. Optical background was recorded with the Stokes beam blocked and was subtracted from all spectra. The background-corrected data were then normalized by shot-averaged nonresonant CARS spectra from argon to correct for the average shape of the Stokes dye laser spectrum. Temperatures were derived from the N_2-containing portion of our CARS spectra using the Sandia CARSFT code [20]. CARSFT computes theoretical CARS spectra and uses a nonlinear optimization routine to find the best fit between experimental and theoretical spectra. The dual-pump CARS convolution equations used by CARSFT are summarized by Hancock *et al*. [21] and only details relevant to our pool-fire spectra are presented here. For thermometry, only the N_2 portion of the spectrum was fit. Fitting parameters included: (1) temperature, (2) a vertical offset that compensated for shot-to-shot variability in optical background, and (3) the N_2 mole fraction, which was used to adjust the relative importance of the resonant to nonresonant CARS signals, as the nonresonant bath-gas contribution varied on a shot-to-shot basis in the turbulent pool-fire environment. The theoretical susceptibility was convolved with both the pump-2 laser linewidth (0.08 cm^{-1}) and the measured instrument function of the detection system. Convolutions with the pump-1 and Stokes laser profiles were not required, as the injection-seeded pump-1 linewidth was much narrower than the probed Raman lines, and because the spectra were normalized by measured Stokes laser profiles.

4. Results and Discussion

Two representative single-shot spectra acquired with the CARS probe volume at the center of the sooting pool fire are shown in Fig. 6, where the square root of the CARS intensity is plotted against the anti-Stokes frequency, $\omega_{CARS} = \omega_1 - \omega_S + \omega_2$. Vibrational Raman *Q*-branch contributions from N_2, O_2, and CO_2 as well as pure-rotational *S*-branch lines of H_2 are observed in the spectra. The quantum numbers for the CO_2 Raman lines are indicated in the upper spectrum in Fig. 6. These CO_2 Raman features are identical to those reported by Lucht *et al.* [22] in their dual-pump CARS measurements (see their Fig. 3). The spectra in Fig. 6 were acquired at nearly the same temperature at 1014 K and 1065 K, but with considerably different gas mixtures present in the measurement volume. The uppermost spectrum in Fig. 6 displays evidence of product gas containing CO_2, which has been mixed with heated air, resulting in a significant O_2 Q-branch signature in the spectrum. No evidence of H_2 is observed in the upper spectrum in Fig. 6, which suggests that the products result from combustion at lean or stoichiometric conditions. The lowermost spectrum in Fig. 6 was obtained in a fuel-rich gas mixture with low relative levels of N_2, as shown by the significant nonresonant contribution to the CARS spectrum revealed by the modulation dip near the bandhead of the N_2 Q branch. The lower spectrum contains N_2, but no evidence of O_2, which strongly suggests that mixing with air and fuel-rich combustion has occurred, resulting in significant amounts of H_2 in addition to CO_2.

Representative fits to single-shot dual-pump CARS spectra are shown in Fig. 7, where the square root of the CARS intensity is plotted against the Raman frequencies of the N_2 Q-branch, as probed by $\omega_1 - \omega_S$. Only the N_2-containing portion of the spectrum is displayed, as fits solely to the N_2 Q-branch were used to obtain the temperature. Relative mole-fraction information can be obtained from fits over the full measured domain of our CARS spectra, but these results are more time-consuming and were not available at the time of this writing. The signal levels in our single-shot spectra were generally sufficient to obtain reliable fits to the data, with the major source of noise resulting from shot-to-shot fluctuations in the broadband Stokes-laser spectrum, which distort the CARS spectral shape and are especially apparent if one compares the smoothness of the experimental and theoretical curves at $T = 1301$ K in Fig. 7. The spectra in Fig. 7 have been chosen to show the evolution of the N_2 CARS signature with increasing temperature. For $T < \sim 900$ K, the temperature is primarily determined from the width of the $v = 0$–1 ground level of the N_2 Q-branch signature, which broadens as the rotational population begins to fill higher energy states. Above 900 K, signal from the $v = 1$–2 "hot band" becomes detectable and the relative heights of the ground-state and hot band, as well as their widths are now both key indicators of temperature.

Histograms obtained from ensembles of over 3500 single-shot pool-fire temperature realizations are presented in Fig. 8. The results are an estimate of the pdf of the pool-fire temperature fluctuations at a single point in the center of the fire plume. The two histograms were acquired from different burns that were conducted on the same afternoon to illustrate the reproducibility of the CARS results and of the quiescent fire plumes established in the FLAME facility. Mean CARS-measured temperatures for the two burns are within 5 K, with the average temperature at the center of the fire plume near 1145 K. A 1-mm-diameter sheathed thermocouple with a ~4-sec time constant was mounted approximately 76 mm distant from the CARS probe volume. The thermocouple indicated a mean temperature near 1210 K for the steady state portion of the burns, within 65 K, or 5.6 % of the mean CARS-measured temperatures; which is quite a favorable comparison given the potential for thermocouple measurement bias resulting from radiant exchange between the probe and its surroundings. The standard deviations of the CARS-measured temperature histograms presented in Fig. 8 reveal a difference of 15% between the two experiments, with some differences in the shape of the histogram observed on the cold-gas side of the mean temperature. The temperature data from both burns display significant fluctuations, which are indicative of the large degree of mixing, which brings cold gas to the center of the fire plume even at only 0.5 diameters above the liquid-pool surface.

The accuracy and precision of our CARS thermometer were assessed over a wide range of temperatures by recording single-shot CARS measurements in furnace-heated air, whose temperature could be accurately measured with a thermocouple. The CARS measurement volume was placed at the center of the open tube furnace, and 100 single-laser-shot CARS spectra were acquired at each furnace temperature setting. The mean of the single-shot CARS temperatures are plotted against the thermocouple standard in Fig. 9, where the indicated error bars represent one standard deviation of the single-shot CARS temperature data. At temperatures greater than 500 K, the CARS mean temperatures agree with the furnace thermocouple to within 20 K or 3% of the thermocouple reading, with the bulk of the measurements well within 1.5%, such that the accuracy of our in-fire CARS measurements is ~2-3%. At temperatures less than 800 K, the precision (single standard deviation) of the CARS results is ±20-30 K, or ±4-5% of the absolute temperature. Above 800 K, the precision degrades to ±40-50 K in terms of the absolute temperature, but the relative precision actually improves a bit to ±3.5-4% for temperatures between 973 and 1400 K.

5. Summary and Conclusion

A dual-pump CARS instrument has been constructed for the probing of 2-m-diameter sooting turbulent pool fires at the Sandia FLAME facility. FLAME provides a unique test bed where canonical, quiescent fire plumes can be established with near-laboratory control, and probed with laser diagnostics of laboratory fidelity. The details of our unique dual-pump CARS facility have been presented. The dual-pump approach enables measurements in sooting fires by positioning the N_2 CARS signature in a spectral region that is free of interference from laser-produced C_2 Swan-band emission/absorption and C_2 Raman features. Temperature is determined on a single-laser-shot basis from fits to the N_2 Q-branch signature observed in the dual-pump CARS spectra, and temperature histograms from two separate pool-fire experiments, employing a 90% methanol / 10% toluene blended fuel, are presented. The pool-fire measurements reveal excellent test-to-test repeatability in the mean (within 5 K or 0.4%) and standard deviations (within 53 K or 14%) of the CARS-measured temperature histograms, and the mean CARS temperatures are within 53 K (5.6%) of a nearby thermocouple probe. The broad, 350-400 K standard deviations of the CARS temperature histograms from the pool-fire experiments reveal that turbulent mixing extends to the center of the fire plume at a height of 0.5 pool diameters. Single-shot CARS-measured temperatures from furnace-heated air reveal an accuracy of 2-3% and a precision of ±3.5-5% at temperatures from 500-1400 K, which spans a significant portion of the observed temperatures in the pool-fire experiments. Our dual-pump CARS spectra also contain Raman features from O_2, H_2, and CO_2, from which we are currently extracting single-shot species mole-fraction data for these species relative to the N_2 molecule. This additional scalar information will provide an indicator of the mixture fraction space on a single-shot basis.

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Figure Captions

Figure 1 – Three-dimensional drawing of the FLAME test bay.

Figure 2 – Digital photograph of the methanol/toluene pool fire.

Figure 3 – Energy level diagrams for simultaneous dual-pump CARS probing of multiple species.

Figure 4 – CARS experimental configuration: the orientation of the pump, Stokes and CARS beams in the folded phase-matching configuration is shown.

Figure 5 – Dual-pump CARS instrument as fielded at the Sandia FLAME facility. The arrangement of the laser and optical system in the west laboratory is shown. Legend is as follows: (BS) beam-splitter; (M) dielectric turning mirror; (M2) mirror periscope; (LC) concave lens; (LX) convex lens; (P) turning prism; (PF) turning prism on fine-adjust mount; (PZ) polarizer; (λ/2) half-wave rotator.

Figure 6 – Representative dual-pump CARS spectra acquired from a sooting methanol/toluene pool fire.

Figure 7 – Representative fits to the N_2-containing portion of single-shot dual-pump CARS spectra obtained from a sooting methanol/toluene pool fire.

Figure 8 – Histograms (pdf) of single-shot CARS temperatures obtained from a sooting methanol/toluene pool fire for two repeat burns.

Figure 9 – Results of single-shot CARS temperature measurements in a tube furnace filled with air. Filled circles represent the mean of 100 single-shot CARS measurements at each tube-furnace setting. Error bars represent the standard deviation of the 100 single-shot measurements.

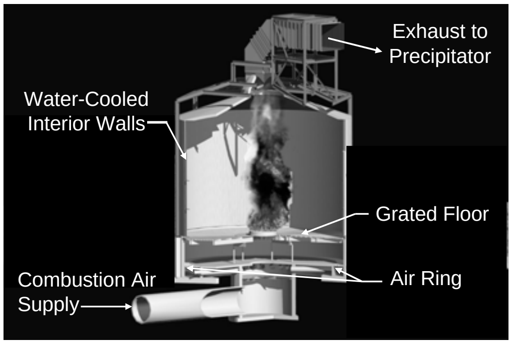


Figure 1

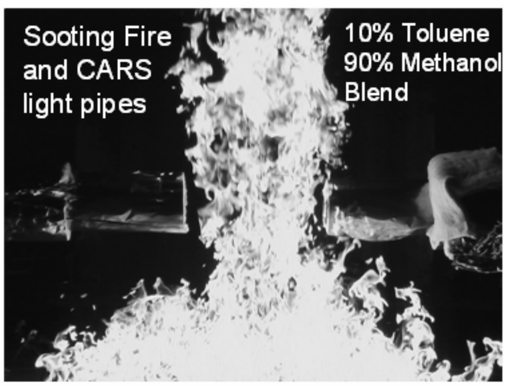


Figure 2

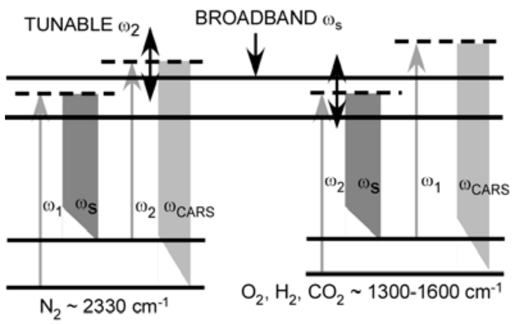
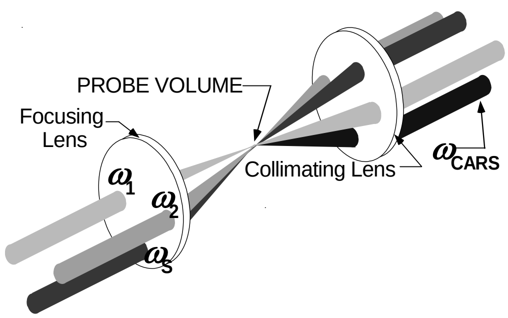


Figure 3



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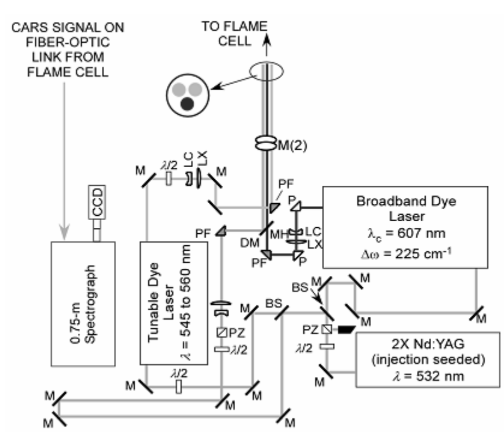

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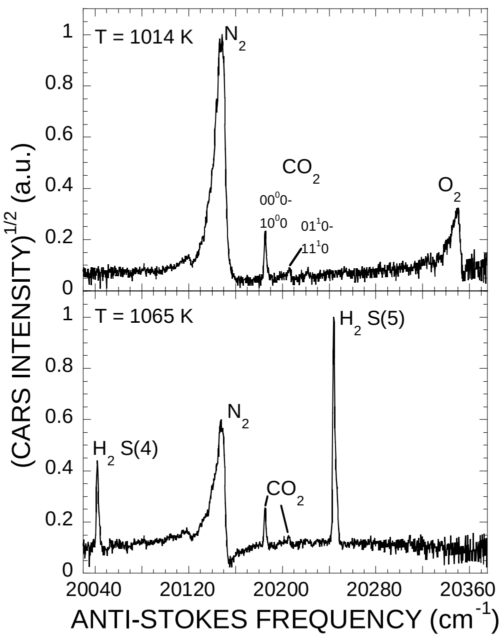

T = 1014 K
N2
CO2
00^0 0-
10^0 0
01^1 0-
11^1 0
O2
T = 1065 K
H2 S(5)
N2
H2 S(4)
CO2
(CARS INTENSITY)^1/2 (a.u.)
ANTI-STOKES FREQUENCY (cm^-1)
0
0.2
0.4
0.6
0.8
1
20040
20120
20200
20280
20360

Figure 6

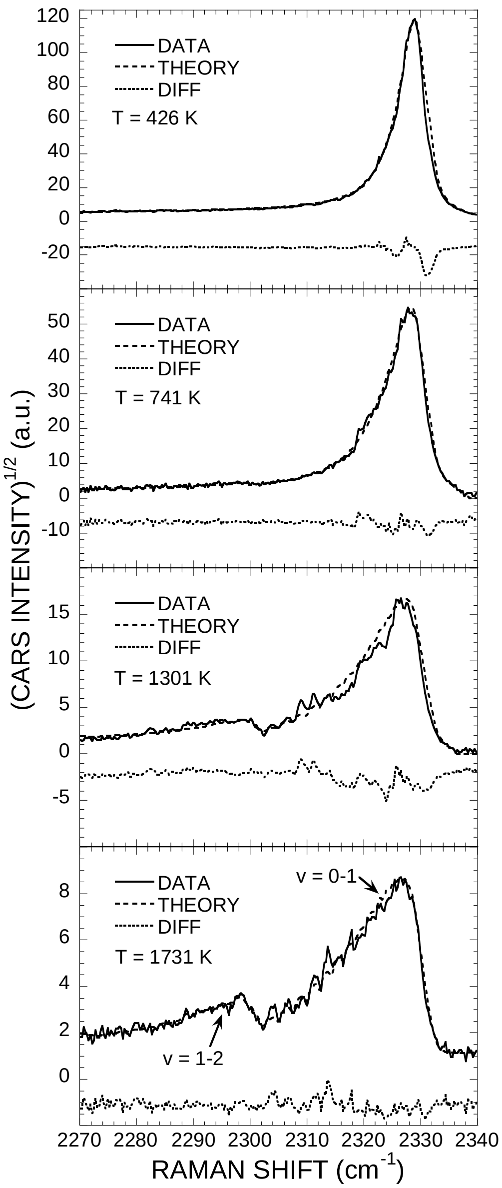
DATA
THEORY
DIFF
T = 426 K
T = 741 K
T = 1301 K
T = 1731 K
v = 0-1
v = 1-2
(CARS INTENSITY)$^{1/2}$ (a.u.)
RAMAN SHIFT (cm^{-1})

Figure 7

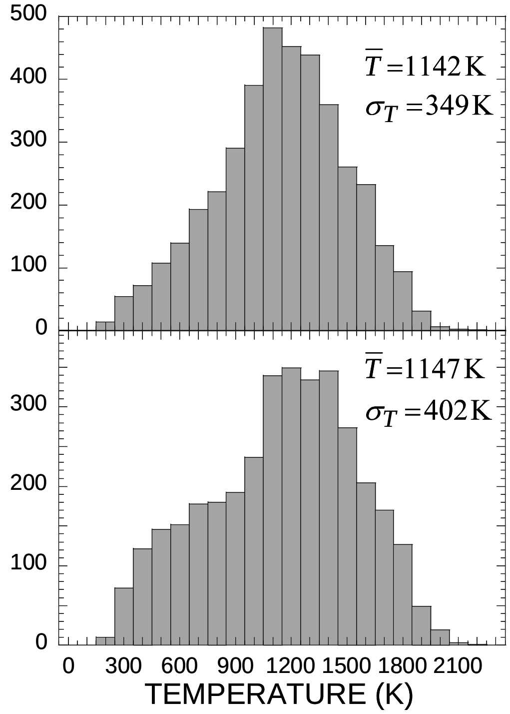

Figure 8

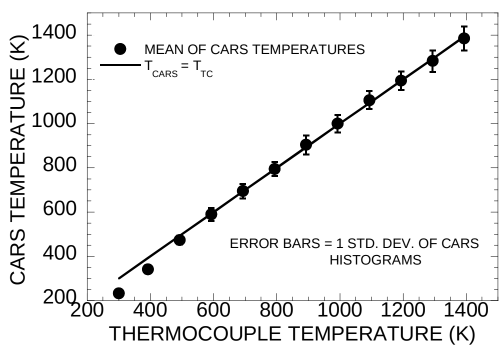

Figure 9