

Multi-Color CARS for Hydrogen-Fueled Scramjet Applications

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Abstract. An approach to CARS, with considerable potential as a diagnostic for advanced airbreathing propulsion devices, is described and demonstrated. The technique is capable of providing simultaneous measurements of the major constituents in H_2 /air combustion. The method employs both broadband and narrowband Stokes beams to generate CARS via two, two-color and two, three-color wave mixing processes. Temperature and concentration information are simultaneously available from N_2 , H_2 , and H_2O . This permits the disappearance of fuel and appearance of product to be monitored. An analysis of the technique's capabilities and future improvements are discussed.

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In the last few years, there has been a rebirth worldwide in hypersonic aerodynamics inspired by visions of sub-orbital global transport, e.g., the Orient Express. Due to aerodynamic heating, flight above a Mach number of five requires cryogenic hydrogen fuel for active cooling of critical aircraft structural components. Furthermore, due to the very high pressures and temperatures which result from slowing the flow to subsonic conditions, the propulsion engines for such aircraft must operate with supersonic combustion, i.e., in the supersonic combusting ramjet (scramjet) mode. Achieving efficient operation in flows moving at speeds greater than 10^3 m/s in combustors of modest length, ≤ 1 m, represents a formidable engineering challenge. Laser diagnostics offer many advantages over more conventional methods in the study of supersonic combusting flows. Measurements can be made in-situ, requiring only optical access. In contrast to physical probing, laser techniques are nonintrusive, and, as such, can be used in environments with extreme temperature or flow conditions without concern for survival. In addition, laser diagnostics are non-perturbing whereas physical probes can affect the combustion and aerodynamic processes being observed, particularly with supersonic flow. Accuracies of laser diagnostic techniques are generally equal to or better than conventional thermocouples and mass

sampling techniques, since corrections are not required to account for factors such as radiation heat transfer or reaction quenching times. Finally, laser diagnostics offer higher spatial resolution since the measurement volumes are determined by the focal volume of the laser beams.

The feasibility of coherent anti-Stokes Raman spectroscopy (CARS) has been demonstrated for supersonic combusting flows [1] and it offers the additional advantage of temporally-resolved static temperature and species concentration measurements. Laser pulse lengths on the order of 10 ns provide measurements which are virtually instantaneous compared to the flow or combustion processes. Since most supersonic combustion test facilities operate for only a short duration, a need exists for simultaneous temperature and multiple species measurements in order to maximize data collection and develop parameter correlations. A CARS technique providing this capability, called dual broadband CARS (DBBC), has been demonstrated in laboratory flames [2] and offers potential for supersonic combustion measurements. Recently, an approach has been developed which is more applicable to H_2 /air combustion, a process being considered for many future SCRAMJET applications as well as being the mainstay of many rocket propulsion approaches. This technique, dual pump-Stokes

CARS (DPSC), is a variation of DBBC offering advantages for this particular combustion process.

This paper describes the demonstration of dual pump-Stokes CARS in a laboratory H_2 /air flame. We begin with an overview of CARS and the various methods for simultaneous measurement of multiple species. Descriptions of the experiment and the apparatus are then included followed by the results. The paper concludes with a discussion of the use of the technique in future supersonic combustion research.

CARS Overview

The theory of CARS and its application as a combustion diagnostic are well explained in several reviews [3–9]. As illustrated in Fig. 1, incident laser beams at frequencies ω_1 and ω_2 (termed the pump and Stokes respectively) interact through the third-order, non-linear susceptibility of the medium, $\chi^{(3)}$, to generate an oscillating polarization and, thus, coherent radiation at frequency $\omega_3 = 2\omega_1 - \omega_2$. When the frequency difference $(\omega_1 - \omega_2)$ is close to the frequency, ω_v , of a Raman resonance in a particular species, the magnitude of the CARS radiation at ω_3 is resonantly enhanced and results in a signature uniquely characteristic of that molecular species. Measurements of medium properties, such as temperature and species concentration, are performed from the shape of the spectral signature, i.e., its intensity variation with frequency, and/or the intensity of the CARS radiation. In airfired combustion, temperature measurements are most commonly made from nitrogen since it is the dominant constituent and is generally present everywhere in large concentration despite the extent of the chemical reaction.

For efficient signal generation, the incident laser beams must be so aligned that the three-wave mixing process is properly phased as shown in Fig. 1. In gases, phase matching is easily satisfied by a collinear ar-

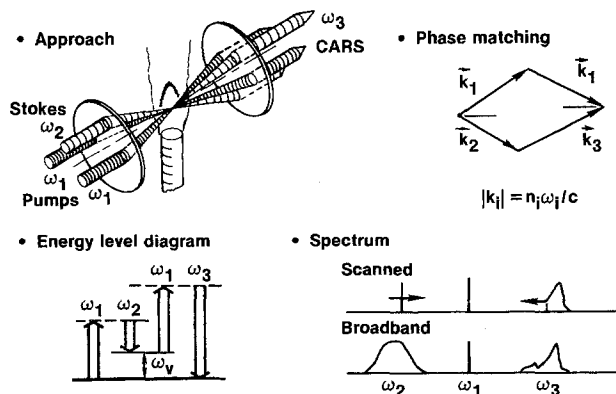


Fig. 1. Coherent anti-Stokes Raman spectroscopy

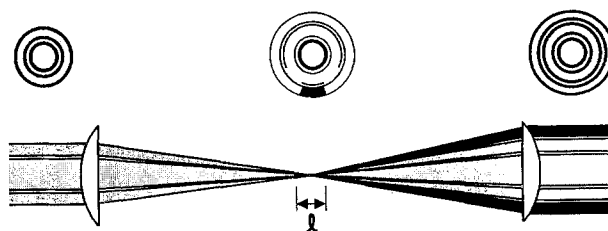


Fig. 2. Unstable-resonator spatially enhanced detection (USED) CARS phase matching scheme

range of the laser beams. In many diagnostic circumstances, however, collinear phase matching leads to poor spatial resolution because the CARS radiation undergoes an integrative growth process. This difficulty can be circumvented by employing crossed-beam phase matching such as BOXCARs (Fig. 1), or a variation thereof. Another phase-matching technique, known as USED CARS, [10–12] makes use of the annular shape of the 532 nm pump beam generated from Nd:YAG lasers with diffractively-coupled unstable resonators. In this approach, the ω_2 Stokes beam is coaxial with and inside of the annular ω_1 pump beam as seen in Fig. 2. The pump and Stokes beams do not intersect until the focal region where the pump beam transforms and fills in. The process may be viewed as the circular summation of many folded BOXCARs (i.e., three dimensional) wave mixing processes around the central Stokes beam resulting in a forward propagating, annular CARS beam outside of the laser beams. This geometry offers greater signal strength at the expense of spatial resolution. The length of the measurement volume corresponds to the length of the focal volume rather than the intersection volume of the three crossed beams. USED CARS offers an advantage in high noise environments because it is less sensitive to vibration and refractive index variations than a BOXCARs phase-matching scheme.

CARS spectra can be acquired in two basic ways as shown in Fig. 1. The scanned approach uses a spectrally-narrow Stokes beam to generate one small portion of the CARS spectrum. The Stokes beam can then be scanned across the frequency range of Raman resonances to generate a high resolution CARS spectrum. This method requires a considerable amount of time and, as such, cannot be used where fluctuations in the medium are occurring or when the event under study is of limited duration. The broadband method overcomes this problem by using a Stokes beam that has a relatively broad spectrum. This allows the entire CARS spectrum to be generated simultaneously, albeit at a loss in signal intensity, allowing virtually instantaneous measurements when short-pulsed lasers are employed. This method is the more useful in examining

most practical combustion systems where turbulence and temperature fluctuations are almost always present.

Multi-Color Wave Mixing

One of the disadvantages of CARS as normally implemented, besides its complexity when compared with spontaneous Raman scattering, is the inability to measure more than a single constituent at a time. Recently, there has been a trend toward multi-color CARS techniques to overcome this limitation [13]. This is motivated by CARS applications to phenomena which are transient or of limited duration. Even for situations where this is not the case, such approaches can greatly expedite data gathering and, more importantly, permit correlations to be developed between the various parameters being measured. In Fig. 3, various approaches to simultaneous multiple species measurements are summarized.

Dual Stokes approaches are a straightforward extension of CARS [14, 15]. For each constituent to be measured, a separate Stokes laser is introduced. Due to the complexity of arranging the beams, generally just two Stokes lasers are used. One performs two separate, two-color wave-mixing sequences to monitor two different constituents. The CARS signatures are located spectrally where they would normally occur, i.e., at the pump frequency plus the Raman shift. In dual pump approaches, [16, 17] two narrowband pump lasers, ω_1 and ω'_1 , are used in conjunction with a single broadband Stokes laser to monitor two species via two separate, three-color wave-mixing sequences. This is merely CARS in its most general form, i.e., three different input waves. In CARS with just two input colors as usually implemented, the two pump waves are frequency degenerate and dual species capability is sacrificed. An interesting aspect of this approach is that the spectra from the two constituents reside in the same

spectral vicinity simplifying optical multichannel detection. The spectra occur at frequencies of $\omega_3 = \omega'_1 + \omega_1 - \omega'_2 = \omega'_1 + \omega_B$ and $\omega'_3 = \omega_1 + \omega'_1 - \omega'_2 = \omega_1 + \omega_A$ and are separated by $\omega_3 - \omega'_3 = (\omega'_1 - \omega_1) + (\omega_B - \omega_A)$. By judicious selection of ω'_1 , spectral overlap can be avoided or minimized to a significant extent.

Both of the foregoing techniques permit measurement of two species simultaneously. They can be extended to a greater number of species by adding more Stokes or pump lasers respectively, but clearly at the cost of increased experimental complexity. An approach, which permits CARS from many species, i.e., at least three, to be generated with no further increase in complexity, is dual broadband CARS [2]. Here, two broadband Stokes lasers are used in conjunction with a pump laser and a combination of two- and three-color wave-mixing processes simultaneously occurs assuming all are phase matched. The two, two-color sequences are normal CARS processes as in the dual-Stokes method described above. However, in addition, there is a three-color wave-mixing process for Raman resonances which correspond to the frequency difference $\omega_2 - \omega'_2$. Since the frequency difference range spanned by two broadband sources is quite large, Raman resonances over a several hundred wavenumber range can be blanketed.

The dual broadband CARS signature resides at the frequency $\omega_3 = \omega_1 + (\omega_2 - \omega'_2) = \omega_1 + \omega_C$ and is the same as would obtain in two-color wave mixing for an ω_C Raman resonance. At the same frequency there is also a contribution from $\omega_1 - \omega'_2$ resonances, i.e., $\omega_3 = \omega_2 + (\omega_1 - \omega'_2) = \omega_2 + \omega_B$. This contribution will be spectrally smeared since it is the broadband ω_2 which is scattering from the excited Raman coherences at $\omega_1 - \omega'_2 = \omega_B$. Depending upon the relative concentrations of the species with ω_A , ω_B resonances, this background may have to be accounted for to perform accurate measurements.

For hydrocarbon-fueled combustion, the spectral location of the various major species resonances is quite fortuitous. The two Stokes sources are positioned to generate CARS from the major products of combustion, namely CO_2 and H_2O . The H_2O bandhead is situated at 3657 cm^{-1} and the major bands of CO_2 at 1285 and 1388 cm^{-1} . Centering the CO_2 Stokes laser near 1326 cm^{-1} permits full coverage of the CO_2 band system. The frequency difference of 2331 cm^{-1} between the two broadband sources permits excitation of the N_2 Raman resonances from which the ω_1 pump beam scatters to generate the CARS signature. Despite the use of two broadband sources in the three-color wave mixing, the spectral resolution is governed, as in the two-color processes, by the spectral width of ω_1 and/or the resolution of the spectrograph employed. This is due to the Raman resonances being well defined

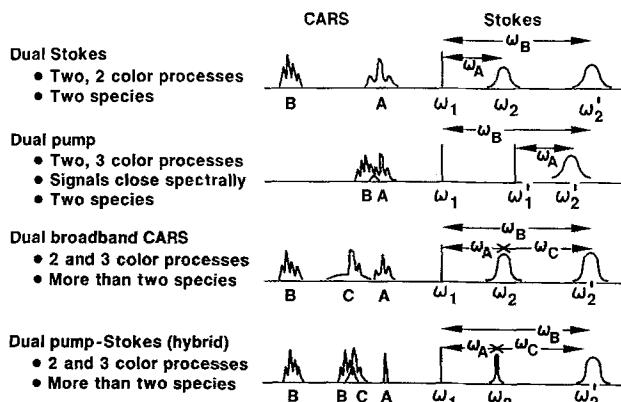


Fig. 3. Multi-color CARS techniques for multiple species measurements

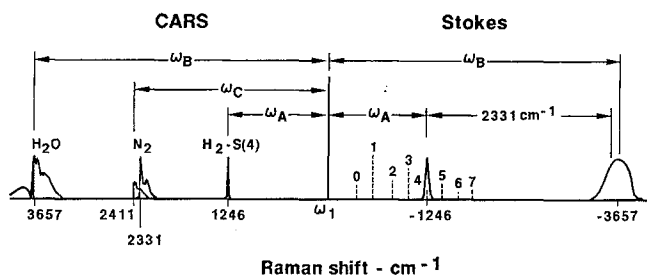


Fig. 4. Dual pump-Stokes CARS approach for H_2 /air combustion measurements

by the specific molecular constituents and not by the manner in which the resonances are excited.

For all wave-mixing combinations to occur in dual broadband CARS, each must be phase matched. There are a number of schemes for doing this including USED CARS [18] and planar or folded BOXCARS or combinations thereof [2]. With any geometry, the three-color dual broadband process will not be as intense as a normal two-color process due to the lower intensity of dye lasers relative to $2 \times Nd$:YAG pump lasers. On the other hand, it will not be as weak as one might suspect due to the spectral integration aspect of dual broadband CARS. That is, there are many frequency combinations driving each Raman resonance. For a resonance centered between two, equally-broad Stokes lasers, all of the energy in each laser is employed in the wave mixing. This is unlike normal broadband CARS where only a thin spectral slice of the dye laser drives the Raman resonance.

The last technique shown in Fig. 3, and the focus of this paper, is a hybrid technique termed dual pump-Stokes due to the fact that the low Raman shift source, ω_2 , can serve as both a pump and a Stokes source. If the low frequency Raman resonance is narrow, the strength of the two-color CARS signal is greatly enhanced by contracting the ω_2 bandwidth. In so doing, this will sharpen the underlying, normally diffuse background spectrum in dual broadband CARS for $\omega_1 - \omega_2'$ resonances producing two well-defined spectral signatures in close proximity as in the dual pump approaches. This situation arises in examining multi-color strategies for H_2 /air combustion. As shown in Fig. 4, one Stokes source is centered at 3657 cm^{-1} to track the appearance of H_2O product in the reaction. Subtracting the N_2 Raman shift, one positions the low Raman shift source at 1246 cm^{-1} to generate H_2 CARS from its pure rotational $S(4)$ transition which is very sharp. This permits the disappearance of the fuel to be tracked. By making this laser narrow to enhance the two-color mixing, an H_2O signature appears along with N_2 in the three-color "dual broadband" process. The H_2O bandhead occurs 80 cm^{-1} to the high frequency side of the N_2 signature, i.e., $(3657-1246)-2331$.

Experiment

A demonstration of the dual pump-Stokes CARS technique was made to determine the best approaches for practical implementation including consideration of a method for simultaneously monitoring all of the CARS signals. Another objective was to evaluate the technique's potential for supersonic combustion measurements. In particular, a study of the CARS signal intensities was required to determine if single shot measurements could be made at one atmosphere pressure.

The experiment consisted of CARS measurements made at several points in a H_2 /air diffusion flame from three of the main constituents in H_2 /air combustion, H_2 , N_2 , and H_2O . Measurements at each point were made sequentially for each species by slewing the spectrometer to the appropriate spectral region. The H_2 /air diffusion flame was generated using a symmetric Wolfhard-Parker burner with gas slots 45 mm long and 5 mm wide for the H_2 and 10 mm wide for each of the two air slots. The H_2 flow was set at 2.8 l/min to establish a convenient flame height and the airflow was adjusted to produce a steady, laminar flame, yielding an overall equivalence ratio of 0.58.

The CARS laser beam axis was aligned parallel to the long axes of the burner gas slots to avoid directing the beams through extreme temperature gradients such as would be encountered in crossing the flame sheets. Also, this aligned the longest axis of the CARS measurement volume (along the laser beam axis) with a uniform region of the flame. A vertical and a horizontal transit of the flame were made and all three spectra were recorded at several points. The peak of the visibly luminescent flame was approximately 55 mm above the burner. The flame sheets converged at about 20 mm and the width of the flame was approximately 12 mm.

The CARS system used for the demonstration is a compact, mobile device, easily configured for a wide variety of CARS experiments. It has been described in detail in [19]. The system consists of three major subsystems; a transmitter, a receiver and an instrument and control rack. The optical system is contained almost entirely in the transmitter and receiver, requiring them to be located at the experimental site. The transmitter is a $95 \text{ cm} \times 156 \text{ cm} \times 97 \text{ cm}$ high cart containing all of the necessary equipment to generate laser beams for producing CARS and directing them in the proper geometry to the measurement point. The optics are located on two pallets, the lower one containing a 20 Hz Nd:YAG laser, two harmonic generators installed in series and the required optics to produce two 532 nm green beams. These beams are directed to the upper pallet by a periscope where they pump the

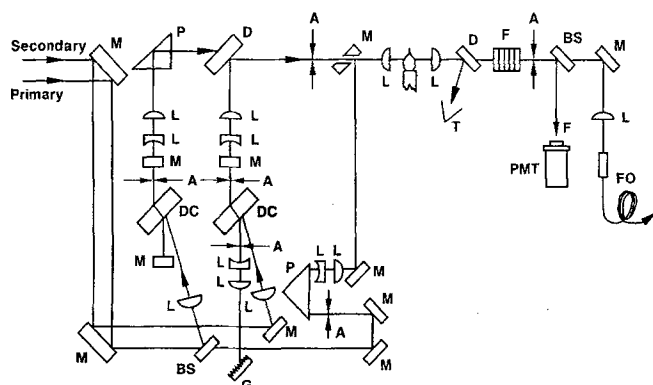


Fig. 5. Optical layout for dual pump-Stokes CARS demonstration. Primary and secondary 532 nm beams enter from the left. Fiber optic cable runs to a remote spectrometer. (Components: A: aperture, BS: beam splitter, D: dichroic, DC: dye cell, F: filter, FO: fiber optic, G: grating, L: lens, M: mirror, P: prism, PMT: photomultiplier tube, T: trap)

Stokes dye lasers and provide the CARS pump beam. The optics on the upper pallet are configured as shown in Fig. 5.

The secondary 532 nm beam and 50% of the primary, more powerful beam are used to pump the two dye lasers. The remainder of the primary 532 nm beam is directed around the pallet, expanded to a 38 mm diameter with a Galilean telescope and provides the CARS pump beam. The ω_2 Stokes laser is a simple, planar, Fabry-Perot dye laser configuration with a 50% output mirror. The dye mixture used is a DCM/DMSO solution lasing at a wavelength of 661.3 nm. The ω_2 laser incorporates a PTR Optics, Inc. TF-17 grating (1200 grooves/mm) as the back mirror to narrow the ω_2 spectrum. To prevent burning of the grating and divergence caused by grating thermal distortion, a Hansch configuration, using a Galilean telescope in the laser cavity, was chosen. The lenses used in the demonstration were uncoated, but, ideally, each surface would be antireflection coated to reduce intra-cavity losses. A large cavity length (32 cm) was established to narrow the ω_2 spectrum as much as possible. The dye mixture used a binary solvent mix of DMSO and methanol in a 50/50 ratio by volume to

shift the Rhodamine 590 dye spectrum to center at 569.7 nm. A small amount of jitter was observed in the ω_2 beam spectrum and, although single shot nonresonant spectra had FWHH's of about 6.7 cm^{-1} , averaged-spectra FWHH's were approximately 7.0 cm^{-1} . FWHH for the ω_2' beam was approximately 350 cm^{-1} .

Both Stokes beams were expanded through $-50/150 \text{ mm}$ telescopes which also provided a means of adjusting the Stokes focal points to coincide with each other and that of the pump beam at the measurement point. The beams were aligned to co-propagate using a dichroic and then aligned along the center of the annular pump beam in a USED CARS phase-matching configuration using a 532 nm mirror with a 12 mm hole at a 45 deg angle of incidence. This geometry provides the simplest method of phase matching all of the two- and three-color wave-mixing processes.

The laser beams were directed out of the transmitter, focused to the measurement point with a 300 mm lens, recollimated along with the CARS beams and directed into the receiver. This subsystem, also breadboarded, contains the optics to separate the CARS beams from the lasers and to focus the signals into an optical fiber. Filtering was done with a 532 nm turning mirror to eliminate most of the laser beams and a filter stack which eliminated the balance of the residual pump and Stokes beams. A clear flat beam-splitter directed about 2% of the CARS signal into a photomultiplier through filters which eliminated any residual pump and Stokes beams. Alignment of the laser beams was optimized by steering adjustments of the Stokes telescopes to maximize the signal on the PMT. The filter stack and PMT filters were changed for monitoring the CARS signals from the different constituents. Table 1 shows the filters used for each spectral region.

The CARS signals are focused into a $50 \mu\text{m}$ core diameter optical fiber with a N.A. of 0.14 for transmission to a remote $f/3.6$ 0.75 m spectrometer located in the instrument and control subsystem. The spectrograph houses a 200 mm dia. concave holographic

Table 1. Receiver filters to observe CARS in H_2/air combustion

Location	$\text{H}_2\text{-S}(4)$ [498.9 nm]	N_2 (473.3 nm)	H_2O (445.4 nm)
Receiver Stack	Corion 500 nm bandpass (8 nm bandwidth)	Corion 550 nm cut-off Ditric 532 nm notch Schott BG 39 (3 mm)	Corion 550 nm cut-off Ditric 532 nm notch Schott BG 39 (3 mm)
Receiver PMT	Corion 500 nm bandpass (10 nm bandwidth)	Corion 470 nm bandpass (10 nm bandwidth) Ditric 532 nm notch	Ditric 500 nm cut-off Ditric 532 nm notch (2 ea)

grating containing 2400 lines/mm. This results in a spectral resolution on the order of 2 cm^{-1} when the CARS signal is spectrally dispersed across the face of the optical multichannel detector (PAR model 1420). A computer records these spectra on magnetic tape for analysis at a later time. A photomultiplier in the spectrometer is monitored when adjusting the fiber optic to the CARS focal point in the receiver to maximize the CARS intensity in the spectrometer.

Results

Representative spectra recorded in the vertical transit through the H_2/air flame are shown in Figs. 6–8. Measurements were taken at 5 mm intervals above the burner surface to a height of 55 mm and an additional measurement was also made at 1 mm. Spectra are presented only to 20 mm above the burner because little change was observed above that height. The CARS signals of the three constituents observed at each point were recorded sequentially, slewing the spectrometer to the appropriate spectral region and installing the correct filters as indicated in Table 1. The spectra shown are averaged over 1200 laser shots (one minute). The average signal count per laser pulse at peak signal intensity is also denoted.

Water vapor spectra (Fig. 6) show the presence of H_2O along the flame centerline, even as low as 1 mm above the burner. Recall that H_2O is probed through conventional two-color wave mixing. The spectral broadening of the bandhead at greater heights in the flame indicates the increasing temperature as the combustion zone is approached. The single peak, to the left of the H_2O spectrum at $\sim 3568\text{ cm}^{-1}$ in the 1 and 5 mm height spectra, is a H_2 vibrational CARS line $v=0$ to 1 (O-branch, $J=3$ to 1, O(3)). Although this is a relatively weak line, the relative concentrations of H_2 and H_2O have made these two signals comparable.

N_2 spectra (Fig. 7), obtained via the "dual broadband" CARS process, show the broadening of the N_2 bandwidth with temperature higher in the flame. The first hot band is apparent to the left of the peak at the 5 mm height and the second hot band begins to appear to the left of the first at 10 mm. A spectrum appears to the right of the N_2 spectrum throughout the flame. This is the three-color CARS H_2O spectrum described earlier as arising from a dual pump process. At the 1 mm height, the H_2 vibrational O(3) line shown in the two-color H_2O spectrum is masked by the N_2 peak in the three-color process. The signal is not as well resolved as the separate, two-color H_2O signature because the former resolution is dependent on the width of the ω_2 Stokes beam. In the two-color process, the pump width is on the order of 1 cm^{-1} while in the three-color H_2O case, the ω_2 beam (6.7 cm^{-1} , viewed

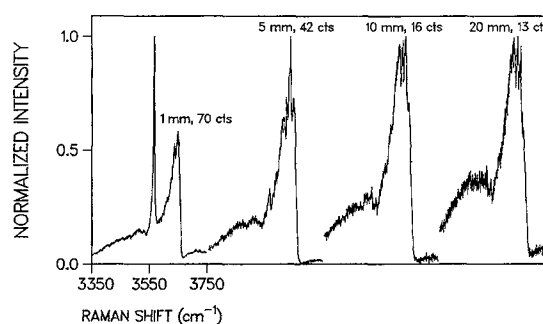


Fig. 6. H_2O spectra in flame. Height above burner and average single shot counts are given. Peak in left-hand plot is the H_2 O-branch $J=3$ line

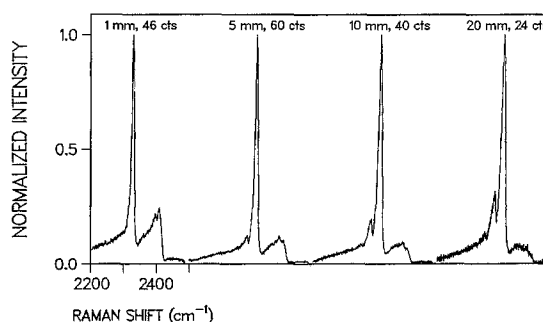


Fig. 7. N_2 spectra in flame. Height above burner and average single shot counts are given. Structure to the right of main peak is H_2O spectrum generated in the three-color CARS process

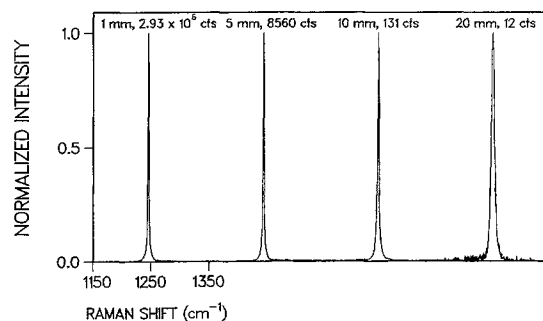


Fig. 8. $\text{H}_2\text{-S}(4)$ spectra in flame. Height above burner and average single shot counts are given

through nonresonant CARS generation) acts as the pump smearing the details in the H_2O spectrum.

From the H_2 spectra in Fig. 8, it is apparent that H_2 is present up to at least 10 mm above the burner. Additional measurements showed its presence up to 15 mm. Beyond this point, the H_2 resonant CARS signal has blended into the spectrally-narrow nonresonant background and it is difficult to distinguish the two. The FWHH of the shot-averaged CARS signature at 1 mm height is 2.0 cm^{-1} . At 55 mm the spectral FWHH is 7.1 cm^{-1} , comparable to that of the nonresonant spectrum (7.0 cm^{-1}), indicating the signal is dominated by the nonresonant background. The FWHH, therefore, can be used as an indicator for the

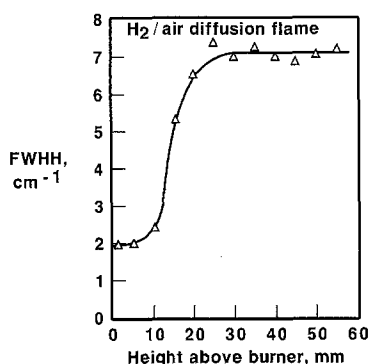


Fig. 9. FWHH of the combined nonresonant and H_2 -S(4) resonant spectrum as a function of height above the burner

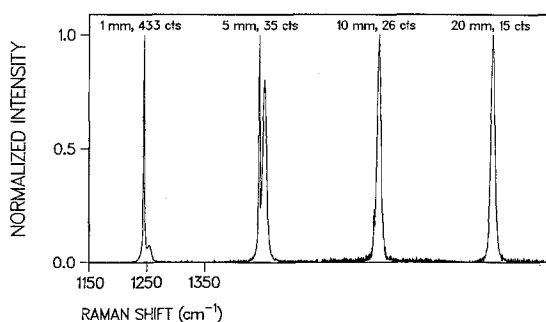


Fig. 10. H_2 -S(4) spectra in flame with Stokes beam displaced to position nonresonant signal 8.5 cm^{-1} from the H_2 -S(4) peak. Right-hand peak is the nonresonant signal. Height above burner and average single shot counts are given

disappearance of H_2 in the flame. A plot of signal FWHH as a function of height (Fig. 9) gives a good indication that the H_2 is virtually depleted by the 20 mm height. Unfortunately this technique probably cannot provide a good quantitative value for H_2 concentration at the spectral dispersion employed, i.e., 0.5 cm^{-1} per pixel. As a result, it will be necessary to separate the nonresonant and H_2 resonant components of the CARS signal near the H_2 -S(4) line. Broadening of the ω_2 dye spectrum would help to distinguish the two signals, but would have a detrimental effect on peak intensity. Another solution would be to shift the Stokes laser to generate the nonresonant CARS signal peak off to one side of the H_2 -S(4) line. Thus, two distinct peaks are generated allowing the resonant and nonresonant portions of the spectrum to be easily separated and simplifying the concentration calculations. Measurements, using this technique from the lower part of the H_2 /air flame on the centerline, are shown in Fig. 10. The Stokes laser was displaced to generate a nonresonant signal 8.5 cm^{-1} from the H_2 -S(4) line. Signal intensity is reduced with this method, however, the degree dependent on how far the nonresonant peak is shifted from the H_2 -S(4) line.

Discussion

The demonstrations here have shown the ability of dual pump-Stokes CARS to make measurements of H_2 , N_2 , and H_2O in a H_2 /air combustion system. As such, the main focus has been on signal strength and the data have not yet been analyzed to determine temperature or concentration. Also, the ability to make simultaneous, single shot measurements must still be improved.

Concern existed initially over the use of the H_2 -S(4) line to monitor H_2 concentration. In the H_2 rotational Raman spectrum shown in Fig. 11, this transition has a relatively low population at low temperatures resulting in a relatively weak signal. However, as the results indicate, the low temperatures occur where the H_2 concentration is relatively high and the CARS signal from the S(4) transition is easily observed. As the temperature rises and the concentration decreases, the population of the S(4) transition increases relative to the lower order transitions reducing the effect of increasing temperature to some degree. As a result, the H_2 -S(4) line is still observed in the higher temperature regions. The lower detectivity level of H_2 has not yet been estimated using this approach, however.

In general, the signal strengths of the spectra were less than adequate for single shot measurements in the demonstrations performed here. Experience has shown that single shot measurements require peak values of at least several hundred detector counts to provide a sufficient signal to noise ratio and to obtain fair accuracy and repeatability. In the hotter regions of the H_2 /air flame, average single shot counts were on the order of 12. In a supersonic combustion environment, signal strength would be lower due to the low static pressures in the combustor. This, however, would be compensated for, to a great extent, by the correspondingly lower static temperatures. As such, efforts to improve the signal by at least an order of magnitude from the current level will be required to reach the single shot threshold.

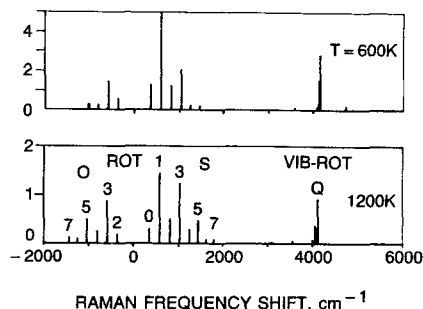


Fig. 11. H_2 Raman spectra at 600 and 1200 K. Stokes beam for H_2 was positioned to generate the nonresonant signal near the S(4) line

A method currently being implemented incorporates a second Nd:YAG laser into the mobile CARS system to increase the available laser power. It will amplify two 1.06 μm beams from the primary laser, eliminating alignment and timing problems involved with the installation of a second, separate laser system. Depending on the saturation level of the lasing medium in the existing laser, the total power available may be more than doubled by the addition of the two amplifiers. In addition, the frequency doubling efficiency will be improved due to the higher power. The 532 nm beams are being split to provide the CARS pump beam and pumps for the ω_2 and ω'_2 lasers reconfigured as oscillator/amplifiers. An increase in the power of each beam by a factor of 3 to 4 will increase the DBBC signal by over an order of magnitude. Additional improvements in signal level can be obtained by increasing the interaction length (e.g., using larger f -number optics or collinear phase matching) and eliminating other losses in the system. These include the use of anti-reflection coatings on all high power optics, elimination of losses in the fiber from the receiver to the spectrometer, spectrometer losses and the examination and correction of other inefficiencies in the system.

Simultaneous acquisition of the multiple CARS signals requires development of a strategy for observing the various signals in a manner which provides all of the information necessary to calculate the temperature and concentrations. Spectral analysis of each signature is required, necessitating the acquisition of all three CARS spectral regions at high dispersion on the optical multichannel detector. From the spectral widths observed in this demonstration, it appears the three regions can be imaged on separate sections of a 1024-channel detector diode array. The N_2 spectrum requires approximately 400 channels and would include the three-color H_2O CARS signature necessary for the data analysis. The H_2O spectrum requires another 400 channels and the narrow H_2 spectrum can be accommodated in the remaining 200 channels. A spectrometer design is required which, when adjusted to the N_2 spectral region, will also direct the H_2 and H_2O spectra onto different sections of the diode array using mirrors and/or cylindrical lenses. Design concepts for such a spectrometer have been discussed in [20] and construction of one such device is presently nearing completion.

The data analysis strategy would begin with obtaining temperature in the measurement volume by analyzing the N_2 spectrum. As shown in Fig. 7, as the temperature rises, the main N_2 peak broadens and "hot band" peaks begin to appear to the left of the main peak. The temperature is derived from signature analysis by either full regression fitting or a variety of

so-called "quick-fit" methods [12]. The H_2O spectrum to the right of the N_2 spectrum will complicate this analysis by raising the N_2 spectrum above the nonresonant baseline. This effect can be handled rigorously using a multi-species CARS analysis. Since the N_2 and H_2O spectra are fairly well separated, a simpler procedure might suffice. Using the two-color H_2O signature, appropriately scaled and spectrally filtered, the H_2O signal can be subtracted from the three-color N_2 signature. This ignores the coherent mixing between the N_2 and H_2O signals which is probably weak in this instance. The accuracy of this approach needs to be investigated through numerical simulation.

Concentration measurements are derived from comparisons of the spectra of the species of interest with the nonresonant background signal [8]. The nonresonant susceptibility is required to make these measurements and must be determined from the spectrally-integrated nonresonant CARS signal acquired in the measurement volume and appropriately referenced to the signal from a reference gas at known conditions. The nonresonant spectrum near the $\text{H}_2\text{-S}(4)$ line provides an excellent source for this information; however, the $\text{H}_2\text{-S}(4)$ spectral line contribution must be removed. With a Stokes laser offset to generate separate resonant and nonresonant peaks (as shown in Fig. 10) or broadened to allow easy distinction between the two signals (possibly about 15 cm^{-1} FWHH), curve-fitting techniques can be used to approximate the shape of the nonresonant spectrum in the region of the S(4) transition. The nonresonant CARS signal of the medium can then be calculated by "correcting" the spectrally-integrated CARS signal generated by the ω_2 Stokes source.

The spectrally-integrated nonresonant reference signal is used to normalize the signal acquired from the measurement volume with respect to fluctuations in ω_1 and ω_2 . The ω_1 and ω_2 beams entering the receiver are split off from the ω'_2 beam and focused into a reference cell with a nonresonant gas such as N_2 at known conditions. The emerging CARS beam is filtered from the laser beams and directed to a PMT. The signal level observed represents the spectrally-integrated nonresonant CARS signal in a medium with a known nonresonant susceptibility and would be recorded with each laser shot using a sample and hold circuit.

To obtain the H_2 and H_2O mole fractions relative to the total nonresonant susceptibility, the total number density is required. This may be obtained in either one of two ways. If the static pressure is known accurately at the measurement location, e.g., from a wall pressure tap, the density is calculable from the gas law knowing the temperature. Or the mixture-averaged nonresonant susceptibility per molecule can be assumed. In a mixture dominated by N_2 , this value

will not change significantly with reaction. H_2 has a nonresonant background 24% lower than N_2 , O_2 35% higher, while the H_2O background is about 1.85 times higher [21]. At stoichiometric conditions, the average nonresonant susceptibility will increase by 30% in going from reactants to products. Taking a value in between results in no more than a 15% error. An even better assumption can be made based upon the local temperature. The H_2 -S(4) signal can then be compared to the nonresonant signal to derive the number density of H_2 molecules in the $J=4$ state. Using the temperature derived from the N_2 spectrum, the Boltzmann distribution can be calculated to obtain the fraction of H_2 molecules in $J=4$ and, thus, the H_2 concentration. The nonresonant susceptibility and temperature can be employed with existing CARS codes and analysis techniques to arrive at an H_2O concentration.

By acquiring spectra on the OMD from each of the three constituents, the need for referencing is minimized and additional PMT systems for signal acquisition are eliminated. The analysis is similar to processes currently in use and would not require a great deal of development. As a result, with the modifications described, a CARS system using the dual pump-Stokes technique could provide a useful diagnostic capability for H_2 /air supersonic combustion.

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