Width-Increased Dual-Pump Enhanced Coherent Anti-Stokes Raman Spectroscopy (WIDECARS)

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WIDECARS is a dual-pump coherent anti-Stokes Raman Spectroscopy technique that is capable of simultaneously measuring temperature and species mole fractions of N_2, O_2, H_2, C_2H_4, CO, and CO_2. WIDECARS is designed for measurements of all the major species (except water) in supersonic combustion flows fueled with hydrogen and hydrogen/ethylene mixtures. The two lowest rotational energy levels of hydrogen detectable by WIDECARS are H_2 S(3) and H_2 S(4). The detection of these lines gives the system the capability to measure temperature and species concentrations in regions of the flow containing pure hydrogen fuel at room temperature.

Introduction

The design of air-breathing supersonic combustion engines is currently being facilitated by creating computational fluid dynamic (CFD) models of turbulent supersonic combustion flow as in Ref [1]. To create and verify CFD models, time-resolved fundamental properties of turbulent supersonic combustion need to be measured. Coherent anti-Stokes Raman spectroscopy (CARS) [2] is a nonintrusive laser-based measurement technique that can measure space- and time-resolved temperature and species concentrations. Studies of hydrogen-fueled supersonic combustion flows were measured with a CARS system design by O'Byrne *et al* [3]. The CARS system developed by O'Byrne *et al*. can measure temperature and species mole fractions of nitrogen, oxygen, and hydrogen. Typically CARS systems can only measure relative species concentrations, but because all the major species present in the combustion flow (except water) were measured, absolute concentration measurements could be made as discussed in Ref. 3.

Future studies are planned to revisit supersonic combustion flow with hydrogen fuel and a mixture of ethylene and hydrogen fuel. To fully characterize these flows, it is desirable to make measurements of as many properties as possible for the full range of conditions within the flow. In these flows, pure, room-temperature fuel is injected into the flow; therefore some regions of the flow will contain only room temperature fuel (hydrogen and or ethylene). The O'Byrne *et al.* CARS system is incapable of making measurements in pure, room temperature hydrogen as discussed in Ref [4]. The O'Byrne *et al*. system is not ideal for measuring combustion of an ethylene-hydrogen fuel mixture because it does not have the capability to measure any of the carbon species present in the flow (CO_2, C_2H_4, and CO).

Others have designed CARS systems that are closer to the desired measurement attributes for these flows. Kearney *et al*. [5] and Beyrau *et al*. [6] have measured many of the major species present in ethylene- and hydrogen-fueled combustion. Additionally, the Kearney *et al.* system is capable of the needed dynamic temperature range in pure hydrogen. Flores [7] designed a CARS system that can measure all major species in ethylene and hydrogen combustion except water and that can measure pure hydrogen from room temperature to flame temperatures. In the present work, a new CARS system called Width Increased Dual-pump Enhanced CARS (WIDECARS) has been designed to improve on the Flores system by using the dual-pump CARS technique [8]. WIDECARS increases the possible spectral resolution, the energy density of the broadband dye laser, and the efficiency of phase matching as compared to the Flores system, while maintaining the same spectral coverage.

WIDECARS is a dual-pump CARS method that employs a broadband dye laser with a FWHM of ~18 nm. This spectral range is twice as wide as a typical CARS broadband dye laser with a gain medium of Rhodamine dyes [3,5,6,8,9,10]. With WIDECARS, all probed species (N_2, O_2, H_2, C_2H_4, CO, and CO_2) are measured simultaneously. This allows for a more complete characterization of the flow. Because WIDECARS measures all the major species (besides water) present in hydrogen-ethylene combustion, absolute species mole fractions can be measured, as demonstrated by O'Byrne *et al*.

To demonstrate the measurement potential of WIDECARS, measurements were made of gas mixtures containing the species WIDECARS was designed to measure. This article includes a discussion of the design of WIDECARS, a comparison of the system's capabilities to those of

other existing CARS systems, a description of the experimental setup of WIDECARS, and the results of measurements made with a WIDECARS system in various gas mixtures.

The Design of WIDECARS

This section discusses how and why WIDECARS was designed. To better understand the capabilities and limitations of WIDECARS, comparisons are made with other similar CARS systems. WIDECARS has been designed to measure mole fractions of all the major species (besides water) of supersonic combustion fueled with either pure hydrogen or hydrogen mixed with ethylene. This design allows WIDECARS to make absolute mole fraction measurements in these types of flows. The major species present in these flows are nitrogen, oxygen, water, hydrogen, ethylene, carbon monoxide, and carbon dioxide. Additionally, it has been designed to be capable of making these measurements at all conditions expected in the flow.

WIDECARS is designed to be a simple extension of the dual-pump CARS technique. The dual-pump technique [8] uses three lasers of different frequencies. This allows for two frequency combinations and therefore, dual-pump CARS simultaneously probes two spectral regions. This design doubles the spectral probing range as compared to a typical system while avoiding the complexity of a fourth laser beam and/or additional or complicated detection system as used in Ref. 10-13.

Figure 1 shows the spectral region probed by WIDECARS (red solid and dashed lines) and compares it to other similar CARS systems. The solid lines show the spectral region probed by the broadband dye laser and Nd:YAG laser frequency combination. For dual-pump techniques, the dashed lines represent the regions probed by the broadband dye laser and narrowband dye laser frequency combination. The probed regions are drawn in the shape of the spectral profile of the broadband dye laser from each CARS system and are plotted as normalized probing energy verses frequency (Raman shift). The broadband dye laser spectrum for each system was created using the dye solutions indicated in the legend of Fig. 1 as the oscillator gain medium. Shown in Fig. 1 by green solid and dashed lines, the CARS system used by Kearney *et al.* could have probed the same species as WIDECARS except carbon monoxide. Although the C_2H_4 v(3) band is in the detection range of the Kearney *et al.* system, it cannot be accurately measured as it is directly overlapped with the first vibrational band of the nitrogen spectrum. The light blue solid and dashed lines in Fig. 1 show the regions probed by the CARS dual-pump system of by Beyrau *et al.* While the Beyrau *et al.* system can simultaneously detect all the same species as WIDECARS, it only detects half as many of the vibrational spectral bands of carbon dioxide. Like the Kearney *et al.* system for the C_2H_4 v(3) line, in the Beyrau *et al.* system the C_2H_4 v(2) band is directly overlapped with the lowest energy nitrogen vibrational band and therefore cannot be measured accurately.

The O'Byrne *et al.* system's region probed by the frequency combination of the broadband dye laser and the Nd:YAG laser is the same as the Kearney *et al.* system. However, its probing region formed by the combination of broadband dye laser and narrowband dye laser frequencies is the same as the Beyrau *et al.* system. Compared to the other dual-pump systems in Fig. 1 the width of the broadband dye laser of WIDECARS is approximately doubled. This halves the energy probing per wave number and thus halves the signal from each spectral line, assuming constant dye laser energy.

In fuel-injected flows which WIDECARS is designed to measure, parts of the flow contain pure fuel (hydrogen or a mixture of hydrogen and ethylene). These fuels are injected at

room temperature and therefore the system must be able to measure in pure fuels at room temperature. In Fig. 2 the square root of the theoretical CARS signal peak heights (signal strengths) of H_2 S(3), H_2 S(4), H_2 S(5) and H_2 S(6) in pure hydrogen are plotted as a function of temperature. The signal strengths of these S branch lines were computed in a CARSFT code that was modified as described in Ref [3]. These S branch spectral lines are the most populated in the probing region of WIDECARS (shown in Fig. 1) for the temperature range in which measurements will be made. Below ~450 K as the hydrogen gas temperature decreases towards room temperature, all these rotational energy levels become less and less populated. This is indicated in Fig. 2 by a decrease in the signal strength below ~450 K.

As the population of the H_2 rotational energy states decrease, the CARS signal decreases and will eventually fall below the measurement limit of the CARS system. This measurement limit is the amount of signal below which the data analysis software (CARSFT) cannot distinguish the signal from the noise. The measurement limit for different CARS systems will vary with the amount of energy used to probe spectral lines and with other attributes of each system such as detector sensitivity and noise, data analysis method, etc. When the O'Byrne $et\ al.$ system was used to make measurements in supersonic combustion flow [4], it showed a measurement limit lower bound of peak height of 3 (square root of the theoretical signal in CARSFT). Square root of the theoretical signal is used in CARSFT because typically species mole fractions scale with the square root of the CARS signal. Using this measurement limit, the corresponding lower bound of temperature of the H_2 S(6) line is indicated by vertical black dashed lines in Fig. 2. The H_2 S(6) line is not populated enough to measure a CARS signal at temperatures lower than 500 K.

Because of this lack of CARS signal at low temperatures, the O'Byrne $et\ al.$ system was unable in Ref. [4] to measure mole fractions or temperature in regions of the flow containing nearly pure hydrogen fuel injected at around room temperature. The lowest rotational lines detected by the O'Byrne $et\ al.$ system are H_2 S(5) and H_2 S(6). In the regions of the flow below 500 K, the one detected spectral line of H_2 S(5) was not enough for the CARS signal analysis software (a modified CARSFT code) [3,14] to distinguish between temperature and concentration.

Figure 1 indicates the hydrogen S branch pure rotational lines detected by WIDECARS and other systems. The Beyrau $et\ al.$ system has the same measurement limitation as the O'Byrne $et\ al.$ system: its two lowest energy H_2 S branch lines detected are also H_2 S(5) and H_2 S(6). The lowest energy rotational H_2 lines detected by Kearney $et\ al.$ are H_2 S(4) and H_2 S(5), which allows for measurements in pure hydrogen down to ~275 K (below room temperature). This is shown in Fig. 2 by the vertical solid black line indicating the lower bound temperature measurement limit of the H_2 S(5) line. However, with this scheme at room temperature, measurements are just above the measurement limit, which may result in noisy data.

The two lowest energy rotational hydrogen lines probed with WIDECARS are H_2 S(3) and H_2 S(4). The inclusion of H_2 S(3) and H_2 S(4) lines not only guarantees concentration and temperature measurement down to room temperature in pure hydrogen, but also allows for hydrogen detection in mixes of room temperature hydrogen and ethylene fuel. In Fig. 3 the square root of theoretical CARS signal peak height at room temperature of hydrogen S branch lines are plotted against hydrogen mole fraction where the balance species is ethylene. Assuming the same lower bound measurement limit as before (square root of the theoretical signal of 3), a black horizontal line is used to indicate the measurement limit in Fig. 3. The H_2 S(4) line height is under the measurement limit for mole fractions less than 0.50. Therefore H_2

S(3) is included in the WIDECARS probing region to allow measurement of hydrogen to concentrations down to 0.03 mole fraction at room temperature.

A spectral or empirical model of the vibrational C_2H_4 v(3) band has not yet been developed for the CARSFT code to be used to analyze spectra measured with WIDECARS. Because of the lack of an ethylene model it is possible that, in regions of the flow containing room temperature fuel mix, the hydrogen rotational lines will be needed to determine temperature. As shown in Fig. 3 to measure temperature the H_2 S(3), H_2 S(4) line combination will be needed to measure temperature for mole fractions of hydrogen below 0.95.

In summary, compared to the other dual-pump techniques featured in Fig. 1 WIDECARS has the potential capability to more fully characterize hydrogen and ethylene fueled supersonic combustion flows. This capability is achieved by increasing the width of the broadband dye laser to include the measurement of all the desired species concentrations and the dynamic temperature range in room temperature fuel.

Shown in black in Fig. 1 is the probing region of the Flores system. This system has the capability to detect the same spectral lines as WIDECARS. The Flores system uses a ~40 nm FWHM dual dye signal Stokes laser to cover the desired spectral region. The Flores system does not use the dual-pump technique and therefore the CARS signal spectral range is the same as the probed region. A feature of dual-pump CARS is that the two spectral ranges probed are overlapped on the CCD camera, resulting in wide spectral coverage simultaneous with higher resolution. A disadvantage of this feature is that spectral lines can overlap, in which case the difficulty of data analysis may be increased. If the same collection setup used for WIDECARS was used for the Flores system, a grating with half the dispersion would be required to disperse the full spectral range of the CARS signal on the CCD. Therefore WIDECARS would have twice the spectral resolution as Flores system. Higher spectral resolution allows for increased accuracy of temperature measurements at temperatures for which only the first energy level of the nitrogen vibrational spectrum is populated.

Another limitation of the Flores system is the broadness of the spectral region covered by the dual dye signal Stokes laser, which reduces the efficiency of phase matching for its full spectral probing region [7]. If phase matching is optimized for the center wavelength of the broadband dye laser, then the efficiency of CARS signal generation decreases with increasing difference from the center wavelength. Therefore, the maximum losses of CARS signal occur for the spectra probed with the wavelengths farthest from the center wavelength of the broadband dye laser. 21% for H_2 S(3) and 14% for N_2 are largest losses of the spectra probed by the Flores system. These losses were calculated using the proportionality of the CARS signal intensity to $\left(\dfrac{\sin\frac{\Delta kl}{2}}{\frac{\Delta kl}{2}}\right)^2$ as defined in Ref [2], where Δk is the phase mismatch and l is the length of the probe volume (assumed to be 1.5 mm). The WIDECARS Stokes beam has half the spectral width and therefore increases the efficiency of the phase matching compared to the Flores system. 4.5% for H_2 S(3) and 3% for O_2 are the maximum losses of signal due to phase mismatch for the spectra probed by the WIDECARS.

However, because WIDECARS uses the dual-pump technique its potential signal strength is decreased by a factor of four compared to the Flores system. But WIDECARS reduces the range of the broadband dye laser in comparison to the Flores system; therefore energy available for probing each line is increased. So overall, WIDECARS has a similar signal strength potential as the Flores system with twice the possible spectral resolution, if the same collection setup is used.

Experimental Setup

The experimental setup of WIDECARS consists of a frequency doubled Nd:YAG laser (Spectra Physics DCR-4) (532 nm), a narrowband dye laser (Spectra Physics PDL-2), and a in-house built broadband dye laser. The Nd:YAG laser serves as the pump energy for the narrowband dye and the broadband dye laser. The Nd:YAG laser used for measurements presented in this paper has a 10 Hz repetition rate and produces 10 ns pulses with an output of 370 mJ. The narrowband dye laser is centered at 556.8 nm with a FWHM of 0.07 cm^{-1}. The broadband dye laser spectrum, shown in red in Fig. 4, is centered at 602.3 ± 0.1 nm and has a FWHM of 17.4 ± 0.1 nm with a range (10%-10% of maximum) of 26.6 ± 0.1 nm as measured with a spectrometer with a resolution of 0.48 nm.

The spectrum of the broadband dye laser is produced by using a mixture of Pyrromethene dyes as oscillator gain medium, a mixture of Rhodamine dyes as the amplifier gain medium, and a spectrally selective optic (thin film polarizer) within the oscillator cavity. In the oscillator gain medium, the concentrations of Pyrromethene dyes dissolved in ethanol are 49.08 mg/L PM 597 and 4.60 mg/L PM 650. This dye mixture produces a spectrum which is shown in green in Fig. 4. A thin film polarizer (part TP2607K060 from Rocky Mountain Instruments) placed inside the oscillator cavity creates a spectrum of the desired shape, shown in orange in Fig. 4. The polarizer is positioned between the back mirror and the gain medium at an angle of incidence of 52 degrees. The thin film polarizer coating is centered at 600 nm. Its transmission curve of unpolarized white light at 52 degrees is shown as a dashed blue line in Fig 4. The polarizer not only helps to shape the spectral output of the laser to the desired spectrum but also rejects the unwanted polarization. The oscillator is pumped by the Nd:YAG laser with a fluence of 0.41 mJ/mm^2. The energy efficiency of the oscillator cavity is 21%. The amplifier gain medium is a mixture of Rhodamine dyes with concentrations of 21.7 mg/L R610 and 8.4 mg/L R640 dissolved in methanol. The amplifier shifts the spectral output of the oscillator ~1 nm to the red producing the spectrum shown in red in Fig. 4. When the amplifier is pumped with fluence of 1.6 mJ/mm^2, the overall energy efficiency is of the laser is 8%.

Further details of this laser and its characteristics will be discussed in a future publication. The spectral output of the laser changes by < 1 nm in shape and location during a testing day (~8 hours).   The spectral output from the PM 650 in the dye mixture slowly decreases after several days whether or not the mixture is exposed to pump light. To maintain the spectral output shown in red in Fig. 4, a small amount of PM 650 concentrate is added to the oscillator solution at the beginning of each testing day.

The three lasers are crossed in folded BOXCARS phase matching regime [2] to create the CARS signal. The energies of the lasers, measured directly before the focusing lens, are 34 mJ from the Nd:YAG, 12 mJ from the narrowband dye, and 17 mJ from the broadband dye. To demonstrate the detection range of WIDECARS, a glass cell 100 mm in length and 1 inch in diameter was centered at the measurement volume. Through the glass cell, room temperature gas mixtures (with known concentrations) were flowed at 0.1 slpm flow rate. These commercially purchased gas mixtures were chosen to contain as many of the species measurable by WIDECARS as possible. The concentrations of the species in the mixtures where chosen to assess the measurement limits. The concentrations in the mixtures were certified to 1% of their measured value to verify the accuracy of the CARS system.

The CARS signal, generated in the glass cell, is dispersed by a ¾ m spectrometer with an 1800 line/mm grating. The CARS signal is then collected on to a 1 inch PixelVision (1100×300

pixel) CCD, producing a detector range of ~20 nm or ~600 cm^{-1}. This range is shown in Fig. 4 by vertical black lines. Black diamonds in Fig. 4 show the locations of the spectral lines in the CARS signal that the frequency combination of the laser produces with this system. The detector range allows for collection of the species probed from O_2 to H_2 S(3) but excludes the probed C_2H_4 v(2), H_2 S(6), and H_2 S(7) lines. After collection on the CCD the background is then subtracted from the signal. Then it is divided by the nonresonant spectrum, collected with argon gas in the glass cell. Finally the square root of the data is taken.

Results and Discussion

A demonstration of species detection capability of WIDECARS was performed by taking measurements of gasses slowly flowing through a glass cell. Sample results from the demonstration are shown in Fig. 5 and Fig. 6. All measurements for this demonstration were taken at room temperature.

The gas measured in Fig. 5 has a composition of 40% hydrogen, 1% carbon dioxide, 1% carbon monoxide, 1% ethylene, and 57% nitrogen. The spectral lines visible in Fig. 5 from right to left are: H_2 S(3), H_2 S(4), CO_2 v(1), CO, C_2H_4 v(3), CO_2 2 v(2) and N_2. This single-shot spectra shows that H_2 S(4) is detectable at 0.40 mole fraction which slightly lower than the theoretical measurement limit of 0.50 mole fraction of predicted in Fig. 3. This spectrum also demonstrates that at room temperature ethylene will be detectable down to a 0.01 mole fraction.

In Fig. 6 another single-shot WIDECARS spectrum is shown measured in a gas mixture with a composition of 3% hydrogen, 20% carbon dioxide, 8% carbon monoxide, 10% ethylene, and 59% nitrogen. The spectral lines visible in Fig. 5 from right to left are: H_2 S(3), CO_2 v(1), CO, C_2H_4 v(3), CO_2 2v(2) and N_2. This gas mixture contains a concentration of hydrogen at the previously predicted measurement limit of H_2 S(3) in Fig. 3. This measurement limit may be accurate: Fig. 6 indicates that the H_2 S(3) is barely distinguishable from the noise. To further verify this as the measurement limit the spectrum must be fit using CARSFT.

Figure 7 shows a single-shot measurement made at room temperature in a mixture of hydrogen and oxygen produced by a water welder and air, detected are the spectral lines H_2 S(3), N_2, and O_2. To prevent an explosion, the measurements of the water welder gases were made in the open air and not in the glass cell. Because these measurements were made in open air the concentrations of the shown spectrum are unknown. Figures 5, 6 and 7 in combination show the all species able to be detected with WIDECARS: nitrogen, oxygen, hydrogen, ethylene, carbon monoxide, and carbon dioxide.

Higher temperature spectra of these gas mixtures were not collected because of the ventilation limitation of the laboratory in which this testing was performed. At higher temperatures there are other spectral lines that WIDECARS can detect that are not shown in these results. These lines include H_2 S(5) and higher energy lines of the vibrational bands of the other molecules. As the temperature of the gas species increase, higher energy levels of the molecules will become increasingly populated.

Because the species' spectra are tightly spaced, when the higher energy levels become more populated spectra of some species will become overlapped. Most species combinations that have potential to overlap, such as N_2 and O_2, C_2H_4 and CO, do not coexist at high temperatures. N_2 and CO_2 2 (v2) spectra coexist at high temperatures and therefore can have spectral overlap of their higher energy levels. This overlap of spectra has the potential to increase the difficulty for the CARSFT code to distinguish between the different species spectra. But

Refs. 13, 15, and 16 mention no difficulties in fitting this spectral overlap, perhaps because the spectral shapes of N_2 and CO_2 2 (v2) differ vastly.

If fits of higher temperature measurements with WIDECARS show that the conflict of the N_2 and CO_2 2 (v2) spectra at higher temperatures is a problem, then a possible solution is to change the placement of N_2 and CO_2 2 (v2) spectra. This can be done by tuning the narrowband dye and the broadband dye to a different combination of wavelengths that will place the N_2 spectra in-between H_2 S(3) and H_2 S(4). This alternative spectral arrangement places the CO spectra at the edge of the broadband dye spectrum and may cause the loss of the carbon monoxide measurement.

Another potential problem with the current wavelength combination is the possible interference between spectral response of the C_2 swan band and N_2 as seen in Ref. [17, 18, 19]. Soot can be present in ethylene combustion. If soot is present in significant quantities, a signal from the C_2 swan band will interfere with the nitrogen vibrational signal. This will cause incorrect temperature fitting as shown in Ref. [19]. If significant amount of soot are present in the flow, this problem could be mitigated by using the same previously mentioned alternative spectral arrangement. Unfortunately, the soot signal is probed by the same wavelength combination as oxygen. So while this alternative arrangement would avoid overlap of the soot signal for all other spectra, oxygen measurement would be confounded.

Conclusion

WIDECARS can detect all major species in ethylene and hydrogen combustion except water. This allows for absolute measurement of species concentrations in these types of flows. WIDECARS's inclusion of H_2 S(3) and H_2 S(4) allows temperature measurement in pure hydrogen for the full temperature range (room to flame temperature) and detection of hydrogen at room temperature down to 3% hydrogen mole fraction. This allows for temperature and concentration measurement in fuel injection zones. The measurement limits of WIDECARS for hydrogen have currently only been approximated using theoretical signal levels from CARSFT and the measurement limit from the O'Byrne *et al.* CARS system. The slowly flowing gas cell measurements did not fully verify the measurement limits of the system for hydrogen at room temperature. The actually measurement limits need to be assessed using more gas mixtures with known small concentrations of hydrogen and fitting the measured spectra in CARSFT.

For WIDECARS to make absolute measurements in ethylene combustion, a theoretical or empirical model for ethylene must be developed and added to CARSFT. Also CARSFT is able to fit up to four species simultaneously but WIDECARS requires the ability to fit six species at once. The data analysis capabilities will need to be expanded to allow for WIDECARS measurements.

WIDECARS has currently only been tested on room temperature gases. Next WIDECARS must be tested for the full range of temperatures in a calibration flame. If the spectral overlap of nitrogen and carbon dioxide causes poor fitting at high temperatures or signal from soot causes interference with the nitrogen signal, a slightly different arrangement of wavelength of the lasers may be needed. WIDECARS has been demonstrated to have the capability to detect nitrogen, oxygen, hydrogen, ethylene, carbon monoxide and carbon dioxide simultaneously, therefore this system has the potential to be very useful for measurements of supersonic ethylene and hydrogen combustion flows.

Acknowledgements

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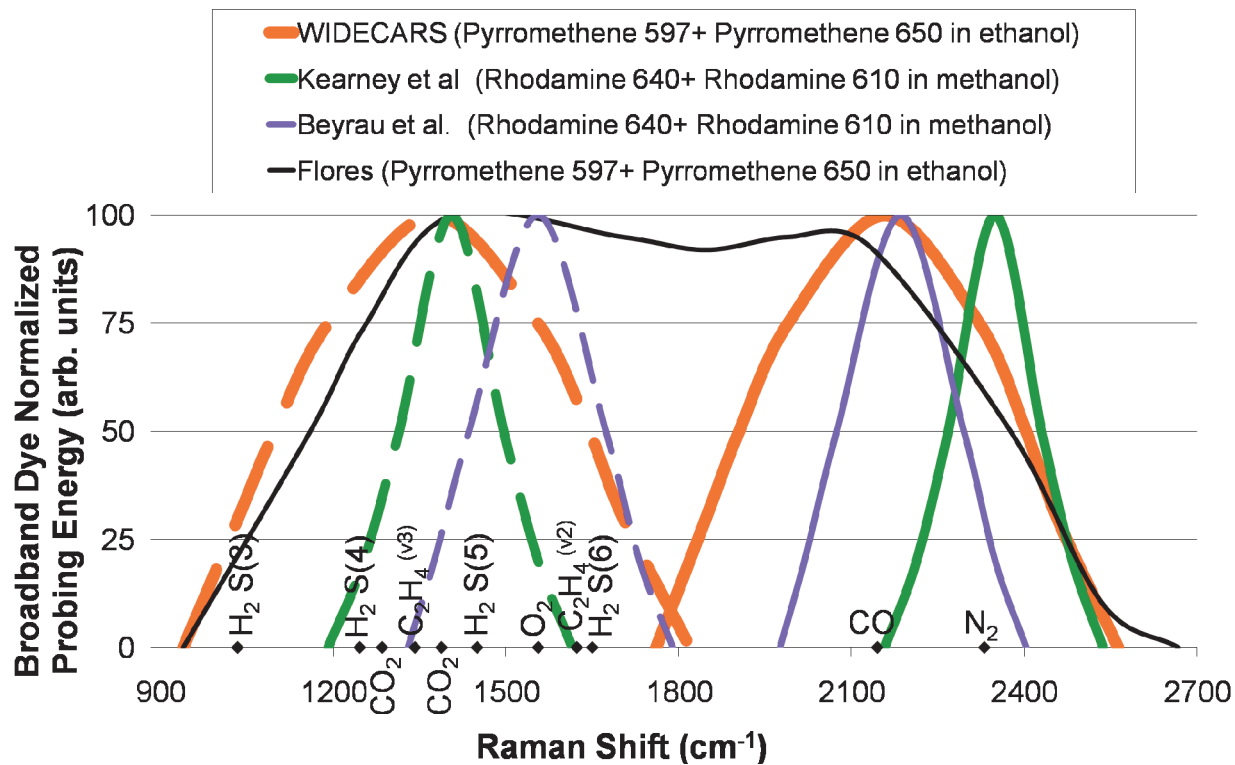
Fig. 1. (Color online) Other CARS techniques spectral probing regions compared to WIDECARS. Solid lines show the broadband spectral regions. Dashed lines represent the additional corresponding dual-pump probing regions. Flores did not use dual-pump CARS.
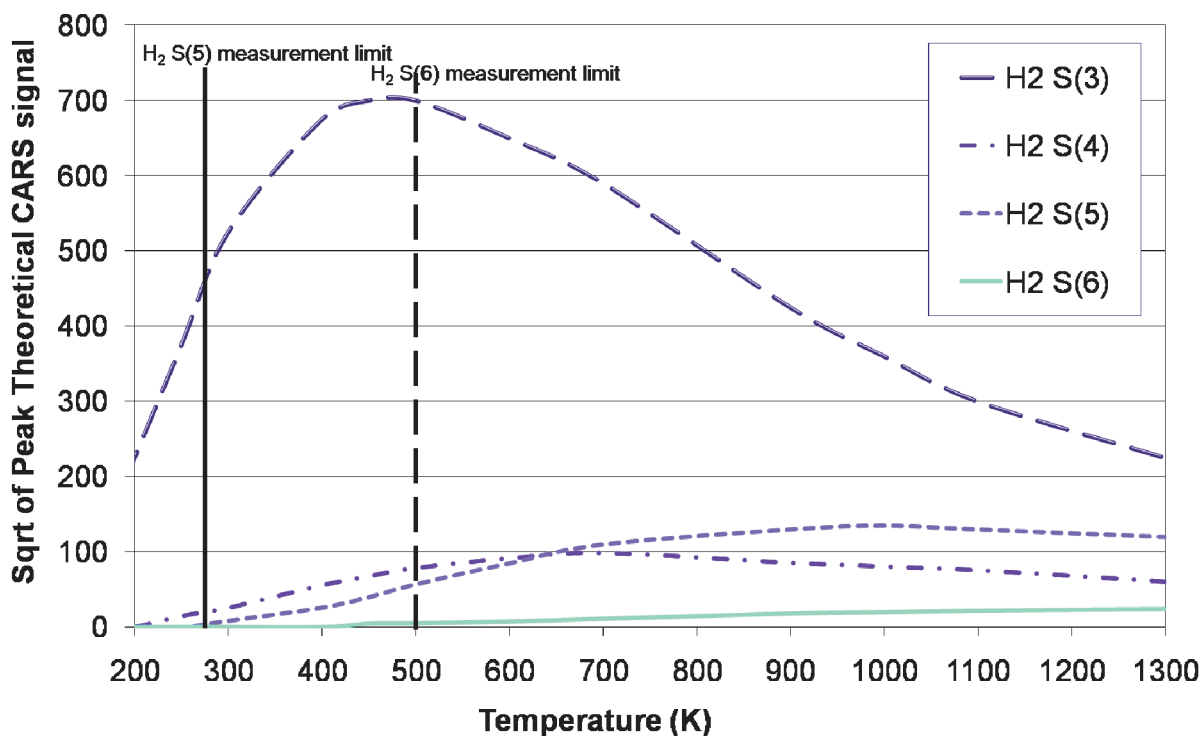
Fig. 2. (Color online) Plot of the square root of the theoretical CARS signal peak height of rotational S branch H_2 lines as a function of temperature. The square root of the theoretical CARS signal peak heights were calculated in CARSFT with 100% hydrogen. The vertical black lines indicate the lowest temperature at which the H_2 S(5) and H_2 S(6) lines can be detected.
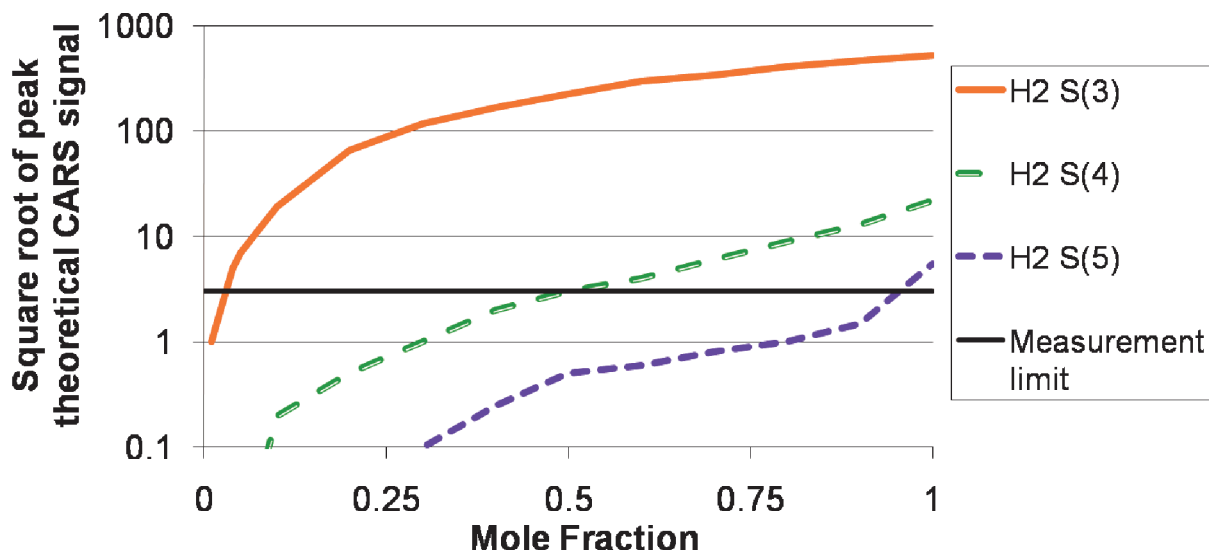
Fig. 3. (Color online) The square root of theoretical CARS signal peak height from CARSFT plotted as a function of the hydrogen mole fraction. This relationship is plotted for H_2 S branch lines at room temperature with a balance gas of ethylene. The measurement limit is a lower bound on the detectable peak height and was obtained from the application of the O'Byrne *et al.* system in a hydrogen fueled supersonic combustion flow.
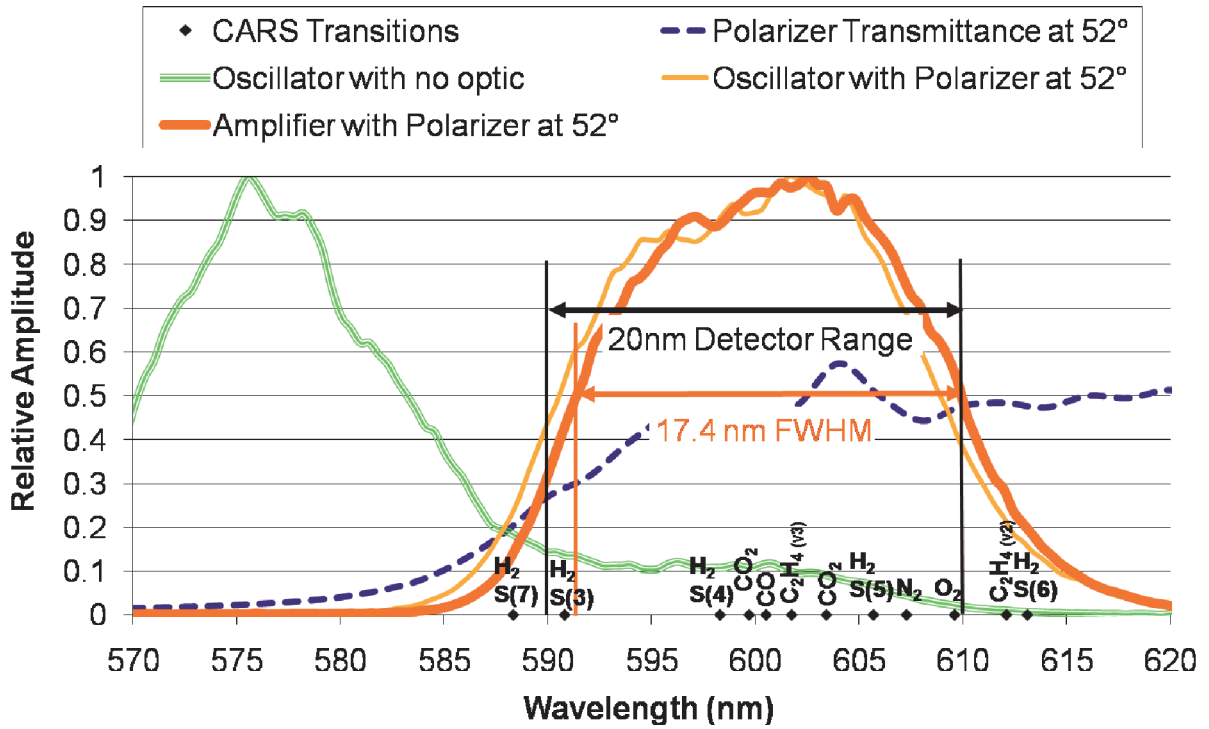
Fig. 4. (Color online) The normalized amplitude of laser output versus wavelength. The green line shows the spectral output of the oscillator of the broadband dye laser with no polarizer in the oscillator cavity. The orange line is the spectral output of the oscillator cavity containing the polarizer. The red line shows the spectral output of the broadband laser after amplification. The polarizer transmittance of unpolarized white light is shown in blue.
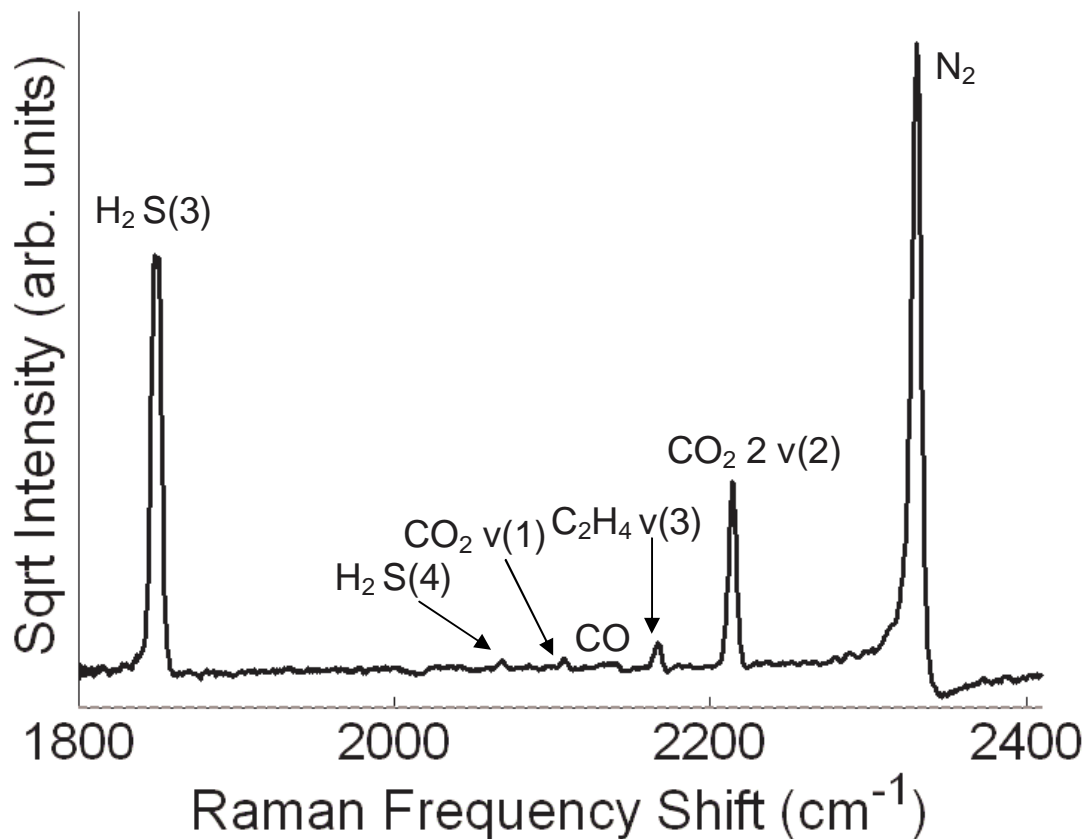
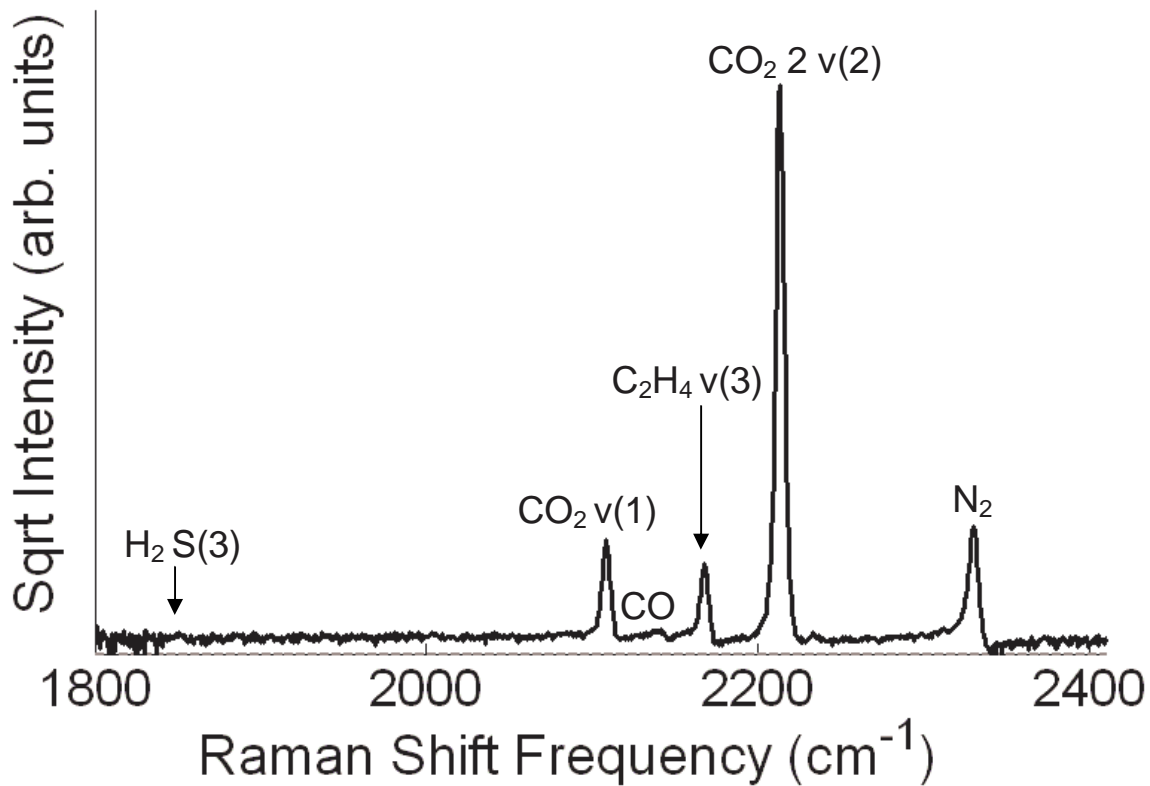Fig. 5. Single-shot CARS spectrum of gas mixture 40% H_2, 1% CO_2, 1% CO, 1% C_2H_4, and 57% N_2.

Fig. 6. Single-shot CARS spectrum of gas mixture 3% H_2, 20% CO_2, 8% CO, 10% C_2H_4, and 59% N_2.
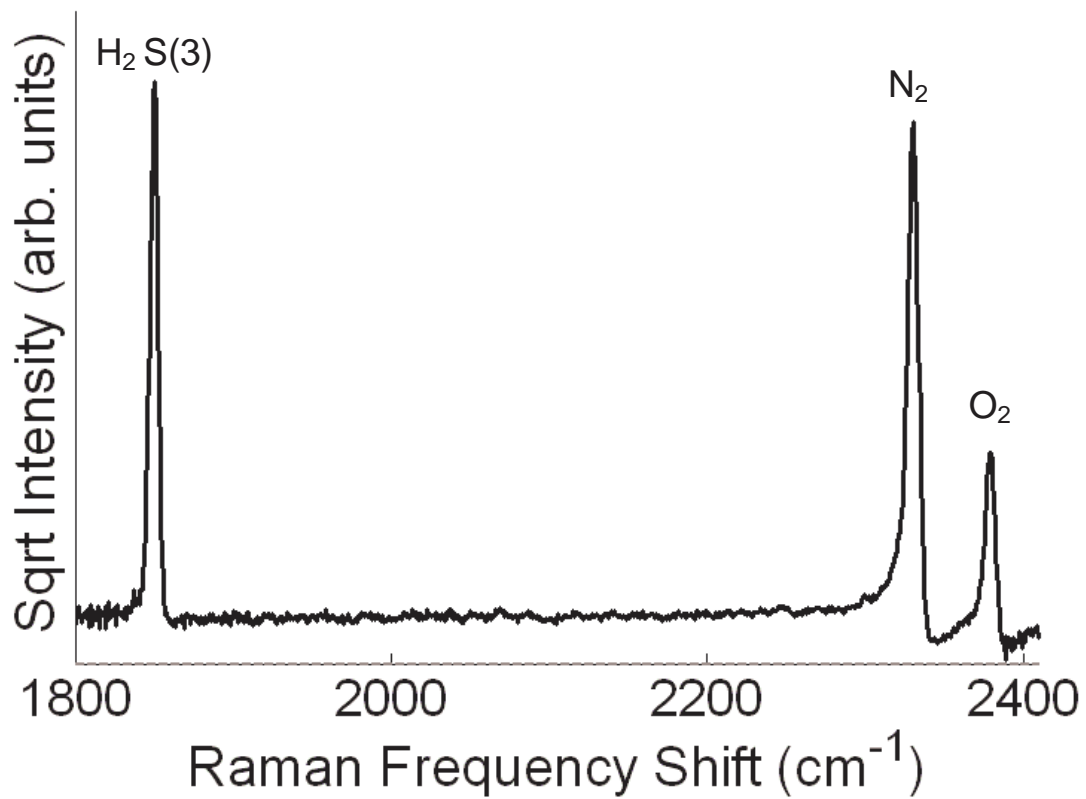
Fig. 7. Single-shot CARS spectrum of unlit water welder gases mixing in air, concentrations unknown.