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Disturbances in N_2-CARS Gas Temperature Measurements in Pulverised Coal-Air-CH_4 Flames

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Abstract

The application of nitrogen vibrational and rotational CARS thermometry to flat, premixed, laminar, methane-air and coal-methane-air flames has been investigated using a Nd:YAG and dye laser system at a low pressure of 30.4 kPa. Although the CARS technique previously has been easily applied to lean and stoichometric methane-air flames, it was found that, in rich methane-air and coal-methane-air flames, C_2 radicals produced by the laser and flame strongly absorbed part of the fundamental band of the CARS spectrum. This imposed severe interference on the N_2-CARS signal. A quantitative investigation of the errors in temperature measurements caused by the C_2 absorption is presented. It is suggested that the effect of C_2 on the CARS signal must be taken into consideration before reliable temperature information can be obtained from rich gas-air flames and coal-air flames.

In this study, a modified experimental and data analysis technique has been applied successfully to eliminate the influences of C_2 on the CARS temperature measurements.

Key words: Laser diagnostics, CARS, C_2 interference, Temperature measurement.

Introduction

Reliable temperature measurements are an important requirement for the understanding of combustion processes and the validation of mathematical models. Although thermocouples have been used successfully to rapidly measure temperatures in combustion processes they have several disadvantages due to their intrusive nature and, in transient systems, due to thermal inertia. Laser techniques offer the means of circumventing many of the disadvantages of physical probes and, for temperature measurements Coherent Anti-Stokes Raman Spectroscopy (CARS) has proved to be reliable and robust in a wide range of combustion systems ranging from laboratory flames through industrial applications to rocket motors. The rapid growth in the application of the technique can be attributed to improved lasers, an enhanced understanding of the CARS process, and the development of sophisticated spectral-synthesis codes, which are necessary for the quantitative interpretation of CARS data.

The CARS technique has been applied very successfully for clean flames. However, in the dirty environment of sooty and coal flames, interpretation of the resulting spectra has proved to be difficult. This is due to particle-induced optical breakdown, which alters the shape and intensity of the experimental spectra and causes a reduction in signal strength by attenuation of the laser and signal beams as they pass through the flame (Beiting, 1986; Luckerath *et al.*, 1995; Hughes *et al.*, 1995). In addition, laser or flame generated C_2 emission, which resides inside the N_2-Q branch spectrum (Singh *et al.*, 1992), disturbs the N_2-CARS signal (Eckbreth, 1979; Beiting,

1986; Boedeker and Dobbs, 1986; Abbott *et al.*, 1988; Bengtsson *et al.*, 1990; D'Alessio *et al.*, 1994; Usta, 1999; Bradley *et al.*, 2001).

Eckbreth (1979) realised that measurements in a highly sooting, laminar propane diffusion flame produced a coherent spectral interference arising from electronic resonance CARS generation from C_2 which was produced by the laser vaporisation of soot. He concluded that a reduction in the Stokes laser bandwidth and use of polarisation filters resulted in low distortion N_2 CARS spectra.

Beiting (1986) found that coal particles induce optical breakdown, creating strong, non-resonant signals. These can be orders of magnitude greater than the nitrogen CARS signals, despite the various inherent filtering processes. The breakdown spectrum is a continuum that cannot be adequately filtered for analytical purposes. One remedy is to reduce the laser focal volume intensity to below the breakdown threshold. However, this can lead to an unacceptable decrease in signal strength. An alternative, used by Abbott *et al.* (1988), is to accept the occasional occurrence of breakdown, but to analyse, in order to obtain temperatures, only "clean" spectra that did not result in breakdown.

In contrast to optical breakdown, Bengtsson *et al.* (1990) concluded that other contributions to erroneous signals that include laser-modulated particle incandescence, fluorescence from large aromatic molecules and Mie scattering from soot are reduced by the filtering techniques that are inherent in CARS. Hence, the main interference is from laser-induced C_2. They analysed only the part of the spectrum that was unaffected by C_2 absorption. They found that single shot temperature errors increased from approximately 2.5% when a whole, 'clean' spectrum was analysed, to 6.0% when only the part of the spectrum that would be uninfluenced by C_2 was considered. In addition, they showed that the resulting temperature was 50-80 K higher with the latter than with the former. Clearly, spectral fitting to only the part of the spectrum unaffected by C_2 absorption compromised the accuracy.

Boedeker and Dobbs (1986) and Bengtsson *et al.* (1990) concluded that in order to measure temperatures reliably in sooty flames, the background generated by C_2 Swan band radiation should be taken into consideration before normal CARS signal processing.

D'Alessio *et al.* (1994) found that, in CH_4/O_2 flames, high molecular mass structures are formed at C/O ratios as low as 0.35. They concluded that below the sooting limit, at which this ratio is 0.45, the C_2 emission arose from photo-fragmentation of high molecular mass structures by the high energy of the laser pulses. Such structures are much more easily photo-fragmented than soot particles.

In the present study, CARS measurements were performed in both pure methane-air flames and coal-methane-air flames at a low pressure of 30.4 kPa to extend the reaction zone thickness and, hence, yield improved spatial resolution. CARS measurements have been obtained very successfully and without difficulty in lean (ϕ=0.6 and 0.7) and stoichiometric methane-air flames. However, in rich methane-air flames (ϕ=1.3) and coal-air–methane flames (for eight different conditions (Usta, 1999)), it was found that the CARS spectra were not only affected by the C_2 emission, but that this was a strong function of the measurement location and the operating conditions. The C_2 excitation arising from the interaction of the laser beam and the flame strongly distorted part of the fundamental band of the N_2 spectrum. Consequently, the effect of the background spectrum generated by C_2 Swan band radiation was investigated by studying the influence on the CARS spectrum and the associated derived temperature of the C_2 background spectrum under a variety of conditions. This paper describes important refinements in the N_2-CARS experimental procedure, acceptance and rejection criteria and data analysis.

Experimental Apparatus

Complete details of the CARS system and associated experimental equipment are presented in Usta (1999) and Bradley *et al.* (2001). Hence, only brief descriptions are given here. The optical arrangement is shown in Figure 1. The Spectron Laser Systems integrated CARS source comprised a Q-switched frequency doubled (512 nm) Nd:YAG laser, with an energy of 350 mJ in 15 ns pulses at 10 Hz and a 35 mJ broad band ($\approx$150 cm^{-1}) dye laser with a centre frequency of 607 nm. The folded BOXCARS phase matching geometry was used due to its good spatial resolution (small measurement volume) and ease of signal collection. The ellipsoidal control volume, 3 mm long, was aligned along isothermal planes, and was 100 μm in diameter. A B& M Spectronic BM100

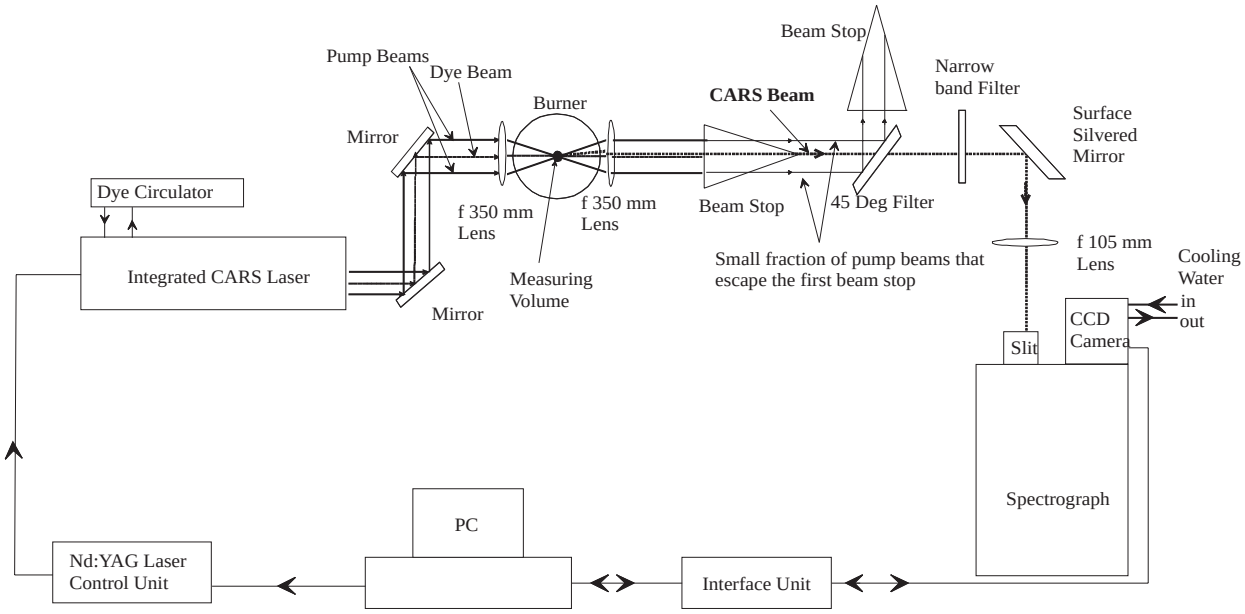

Figure 1. CARS system.

Spectrograph was employed, with a CCD camera, a controller (Model ST-120) and associated software supplied by Princeton Instruments Inc. Nitrogen spectra were analysed with the AERE Harwell computer codes CARP-PC and QUICK to produce single shot temperature measurements.

The CARS system was employed to measure temperatures in a coal flame that was confined within a low pressure. This burner had optical access through two windows, each 60 mm in diameter, which were mounted on opposite flanged openings. An important improvement in the experimental set-up was to provide good optical access to the coal flames while minimising contamination of the windows by coal particles. For this, two manually operated shutters were fixed to the inside of the windows. The shutters were opened only for a short period of time for each measurement. This allowed adequate window cleanliness to perform a complete set of measurements before cleaning was required (Usta, 1999; Bradley *et al.*, 2001).

CARS Technique and Data Analysis

The accuracy of the temperature measurements attainable by CARS is dependent upon the sensitivity of the medium under investigation to changes in temperature. Temperature measurements are derived from the spectral shape of the CARS signature and this is usually quite temperature sensitive. Figure 2 shows the effect of temperature on the normalised spectrum shape for nitrogen at 30.4 kPa. It was computed by the computer program CARP-PC for the temperature range between 300 and 2100 K, in increments of 300 K. At low temperatures, only the ground vibrational state ($\nu = 0 \rightarrow \nu = 1$) and the first few rotational states are significantly populated. As the temperature increases, the increased rotational population broadens this narrow fundamental band, and at about 1000 K the first vibrational 'hot-band' ($\nu = 0 \rightarrow \nu = 2$) becomes significantly populated. The intensity of the hot-band becomes more pronounced and displays a more rotational structure as the temperature increases. In general, there is considerably more information in spectra at high temperatures than in those from cold gas, but the rate of change of spectral shape with temperature is highest at low temperatures (Greenhalgh *et al.*, 1983). An experimental CARS spectrum is a function of the probed medium (N_2) and also of the spectrum of the incident lasers and of any background radiation.

After optimisation of the laser beams and CARS signal, the first stage in the analysis of a clean CARS signal is to remove the effects of the lasers and background radiation. First, the spectrum due to background radiation, without a flame, due to room lights and any laser beam reflection, was recorded by blocking off the Stokes beam in room air. Next, the non-resonant spectrum, which is an accurate representa-

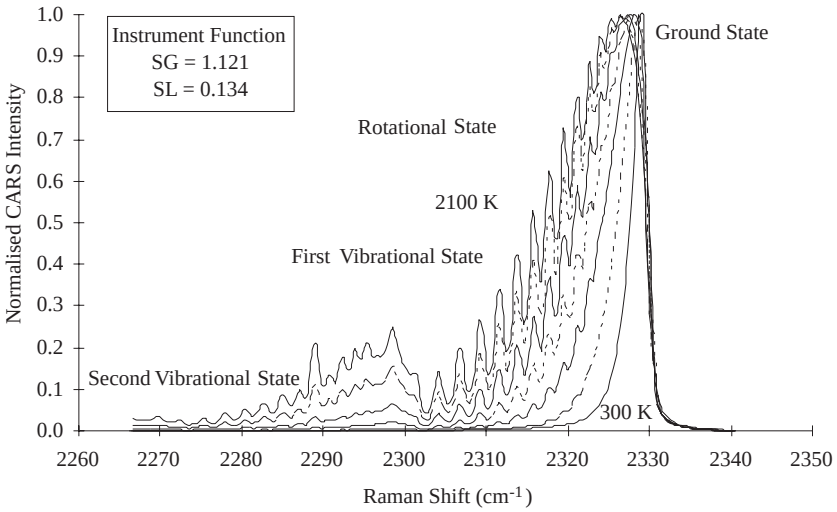

Figure 2. Effect of temperature on the normalised shape of the N_2 CARS spectrum. Curves computed by CARP-PC (from 300 K to 2100 K in increments of 300 K at 30.4 kPa).

tion of the laser spectrum, was obtained byfilling the control volume with CO_2 such that a spectrum was obtained without the presence of N_2. Finally, a CARS spectrum was obtained in room air and, after subtracting the background spectrum, and dividing by the non-resonant spectrum, this was compared with a predicted spectrum to determine any distortion due to the experimental set-up (the instrument function). Finally, the desired gas temperatures were measured in flames. Typical background, non-resonant and CARS spectra are shown in Figure 3, in which the x-axis is the pixel number on the CCD and is a function of the Raman shift.

Temperature information from the experimental spectrum was obtained by comparing measured spectra, after subtracting a background spectrum and normalizing it by a non-resonant one, with a library of theoretical spectra, which were computed as functions of temperature and included the influence of the instrument function.

The experimental spectrum in Figure 3 is reproduced in Figure 4, together with the best-fit theoretical spectrum and the difference between the two.

Modifications to the experimental procedures and data analysis due to C_2 emissions

In rich methane-air and coal-methane-air flames, the background level of the CARS signal is strongly influenced by C_2 emission which is a function of location within the flame. The C_2 emission strongly absorbs part of the fundamental band of the CARS spectrum and, hence, its effect on the CARS spectrum also is a strong function of location within the flame and operating conditions. Therefore, in the present work, it was necessary to record background spectra at every position within the flame for which CARS spectra were required. Temperature information from the experimental spectra was obtained by comparing them, after subtracting individual background spectra and normalization by a general non-resonant one, with a library of theoretical spectra, which were computed as functions of temperature and included the influence of the instrument function.

Because C_2 has an important effect on CARS spectra, it was necessary to determine if interactions between the lasers and the flame might produce more C_2 than would naturally occur. D'Alassio *et al.* (1994) implied that this might occur in rich flames due to photo-fragmentation of macromolecular structures by laser beams even for flames that are under the sooting limit. In the present work, no such interaction was observed in rich methane-air flames (ϕ = 1.3, C/O = 0.244) which were below the sooting limit. This was deduced from the spectrum resulting from the passage of the laser beams (any combination that did not generate a CARS spectrum, e.g. two pump beams, one pump and one Stokes beam or one Stokes beam) through the flame. No change in the resulting background spectrum and hence, C_2 emission, was observed at different locations within the flame.

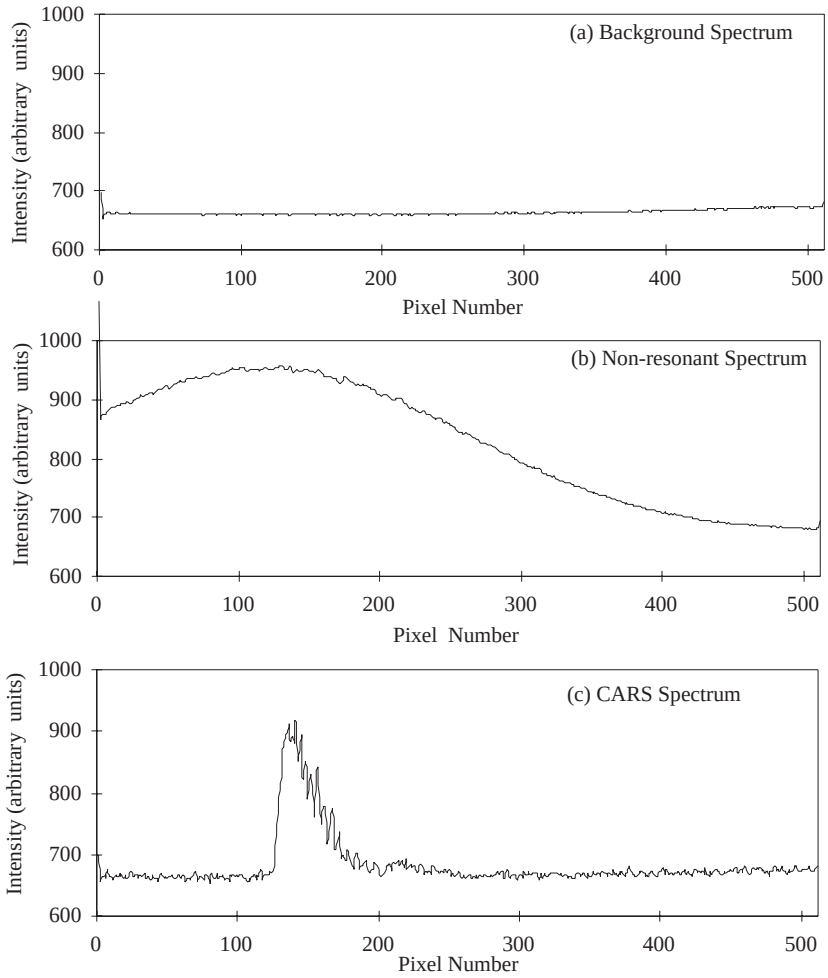

Figure 3. Example of background, non-resonant and CARS spectra (CH_4-air flame, P = 30.4 kPa, $\phi = 0.7$, T = 1770 K).

However, this was not the case in coal studies. C_2 emission was significantly affected by the interaction between laser and soot/particles in the flame. Therefore, the effect of the C_2 influenced background in the present coal-methane-air flames was investigated for background spectra at a variety of conditions.

Figure 5a shows a typical CARS spectrum from the burned products 3.4 mm downstream of the start of the reaction zone of a coal-air-CH_4 flame with an overall equivalence ratio of 2.105 (initially 4.08% CH_4 by volume), a pressure of 30.4 kPa, and an initial temperature of 298 K. Background spectra, which included the C_2 contribution, were also obtained under similar conditions by blocking out, alternately, the pump and dye beams in order to investigate the effect of coherent interference (Bengtsson *et al.*, 1990). Shown by the dashed line in Figure 5b is a background spectrum obtained by blocking off one of the green pump beams. It was obtained under the same conditions and location as the CARS spectrum in Figure 5a. Similarly, the dotted and chain dotted backgrounds in Figure 5b were obtained by blocking off both pump beams and the dye beam respectively. Clearly, the laser induced C_2 intensity depends upon the power of the laser beam(s), which was 30 mJ for the pump beams and 12 mJ for the dye beams. The peaks in the emissions spectra in Figure 5b, at a pixel number of 170, corresponds to a wavelength of 473.83 nm, which is that of C_2. D'Alessio *et al.* (1994), in their studies of sooting flames, also observed this. The background spectrum, measured by blocking the less powerful dye beam, was subtracted from the CARS signal to give the N_2 spectrum since

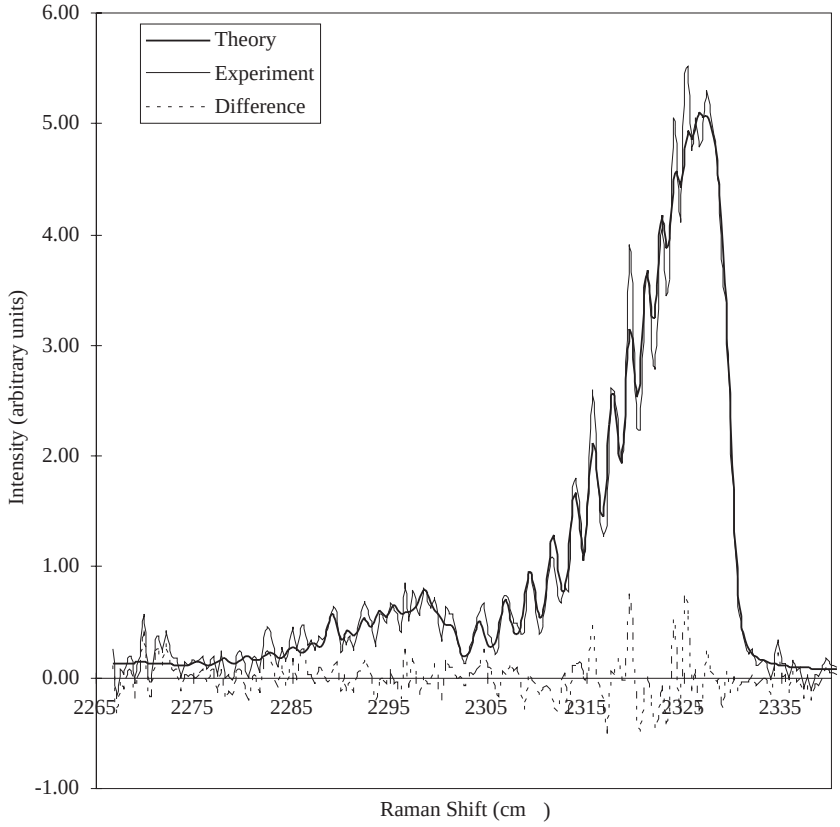

Figure 4. Comparison between the experimental CARS spectrum in Fig. 3 with a fitted theoretical one at 1770 K. The signal to noise ratio is 5.18.

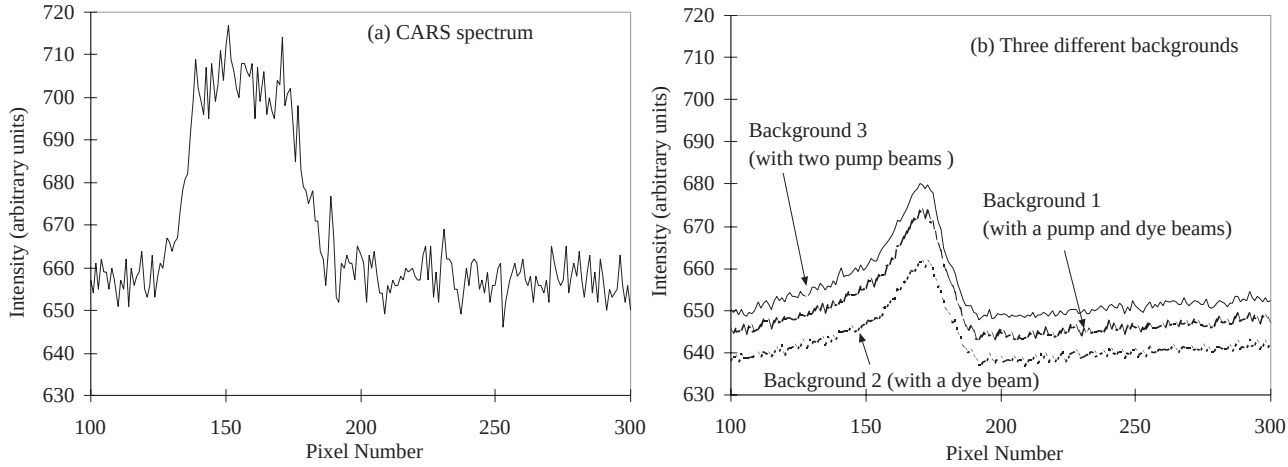

Figure 5. (a) CARS spectrum and (b) Three different backgrounds at the same location in a coal-air-methane flame.

this most accurately represents the induced C_2 contribution within a CARS measurement. It was necessary to measure the background spectrum for every location and every experimental condition. Clearly, shot to shot variations in laser power will result in shot to shot variations in the amount of laser induced C_2 emission. However, since it was not possible to measure the background spectrum simultaneously with a CARS spectrum, only averaged (at the given location and conditions) background spectra were utilised and shot to shot variations were ignored. This was assumed to be acceptable since

intensity, but not spectral shape, was not a function of laser power.

Figure 6 shows the variation in background spectra with location within the coal-methane-air flame discussed above. The variation in spectral shape, probably due to the effect of C_2 emission, is clear. Although this complicates CARS analysis, it can be used to advantage elsewhere because it yields, at least qualitatively, a measure of the concentration of C_2 in the flame.

The peak C_2 emission signal from a methane-air flame and from a coal-methane-air flame are compared in Figures 7a and b. In the methane-air flame, the C_2 emission intensity increases in the reaction zone and then decreases to zero. However, in the coal flame, the C_2 emission in the post-flame zone does not decrease to zero.

Shown in Figure 8 is a raw CARS spectrum obtained within the coal-methane-air flame, a background spectrum obtained in room air and a background spectrum obtained at the same conditions as those for the CARS spectrum. The raw spectrum of Figure 8a was processed using each of the background spectra of Figures 8b and c. Shown in Figure 9 are, for the experimental spectrum in Figure 8a, CARS fitted spectra and the difference between the theoretical and experimental ones when each background in Figures 8b and c is applied. Figures 10 and 11 show similar figures to those of Figures 8 and 9 for a rich methane-air flame ($\phi = 1.3$). The importance of obtaining a specific background spectrum, at the same location and conditions as the CARS measurements, is clear from the much improved fit when that background is utilised.

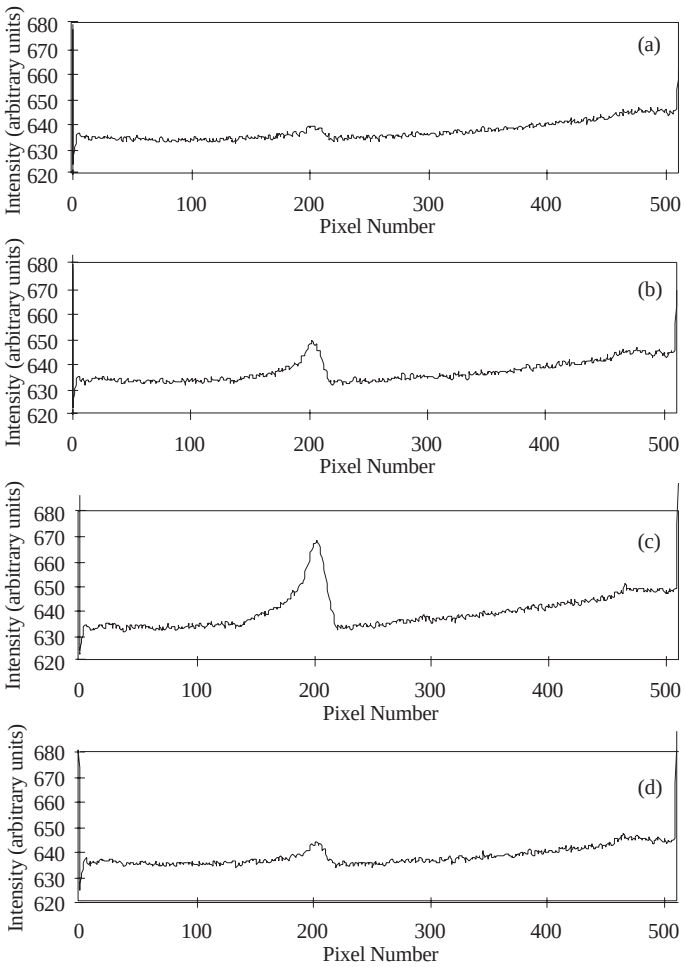

Figure 6. Effect of C_2 emission on the background spectrum (generated using two pump beams) in a coal-air flame at (a) 0.8 mm, (b) 1.9 mm, (c) 7.9 mm, (d) 21.9 mm away from the burner matrix (P = 30.4 kPa, ϕ_{ov}= 2.276, $\phi_{gas} = 1.273$).

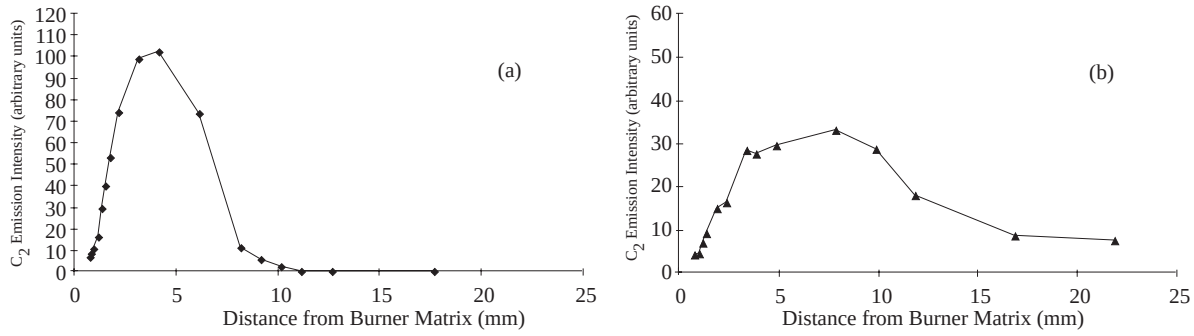

Figure 7. Variation in peak C_2 emission intensity with location in a (a) methane–air flame (P = 30.4 kPa, $\phi = 1.3$) and (b) coal-methane-air flame (P = 30.4 kPa, $\phi_{ov} = 2.276$, $\phi_{gas} = 1.273$).

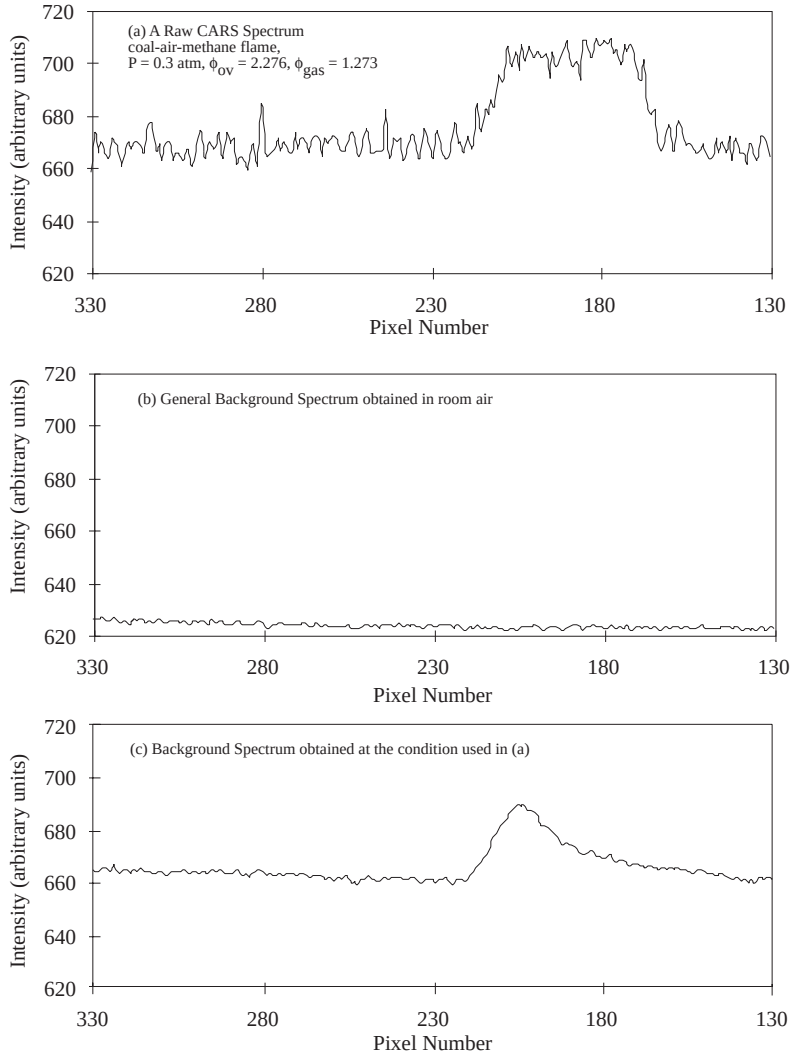

Figure 8. Spectra from a coal-air-methane flame at a pressure of 30.4 kPa, $\phi_{ov} = 2.276$, $\phi_{gas} = 1.273$. (a) Unprocessed CARS spectrum, (b) Background obtained at atmospheric conditions, (c) Background obtained at the same conditions as the CARS spectrum in (a).

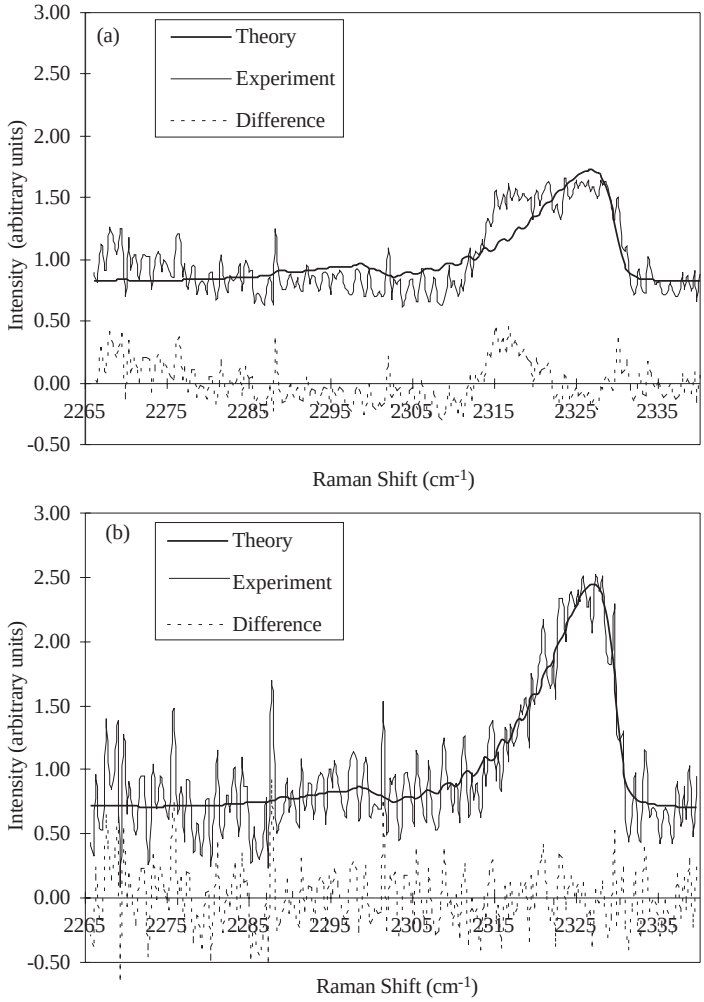

Figure 9. Theoretical curve fit of the CARS spectrum in Fig. 8a. (a) Using the room air background spectrum of Fig. 8b, and (b) using the background spectrum of Fig. 8c.

Although it is not yet possible to experimentally or theoretically quantify the improved accuracy produced with the above methodology, valuable insight may be gained by a comparison with modelled data. Figures 12 and 13 show comparisons between modelled predictions (Usta, 1999; Bradley *et al.*, 2001) and CARS temperature measurements for a rich methane-air flame and methane-coal-air flame respectively. Results of CARS analysis based on the previous method of using a single, general, background spectrum, and one using individual backgrounds are presented. Clearly, good agreement between theoretical and experimental spectra, and reliable measurements of temperature are obtained only when a CARS spectrum is associated with a background one that was obtained under identical conditions.

Acceptance and rejection criteria

Measured and theoretical spectra are unlikely to precisely agree in CARS analysis. In many situations agreement can be very poor due to a number of effects that include shot to shot variations in the spectral distribution of dye and pump laser powers, inhomogeneities in temperature and N_2 concentration within the measurement volume and particles in the flow. Image persistence on the CCD camera is another source of noise but this probably is a small contribution to the total noise. It is usual to reject poor quality experimental spectra that are not well matched by a theoretical one (Bengtsson *et al.*, 1990).

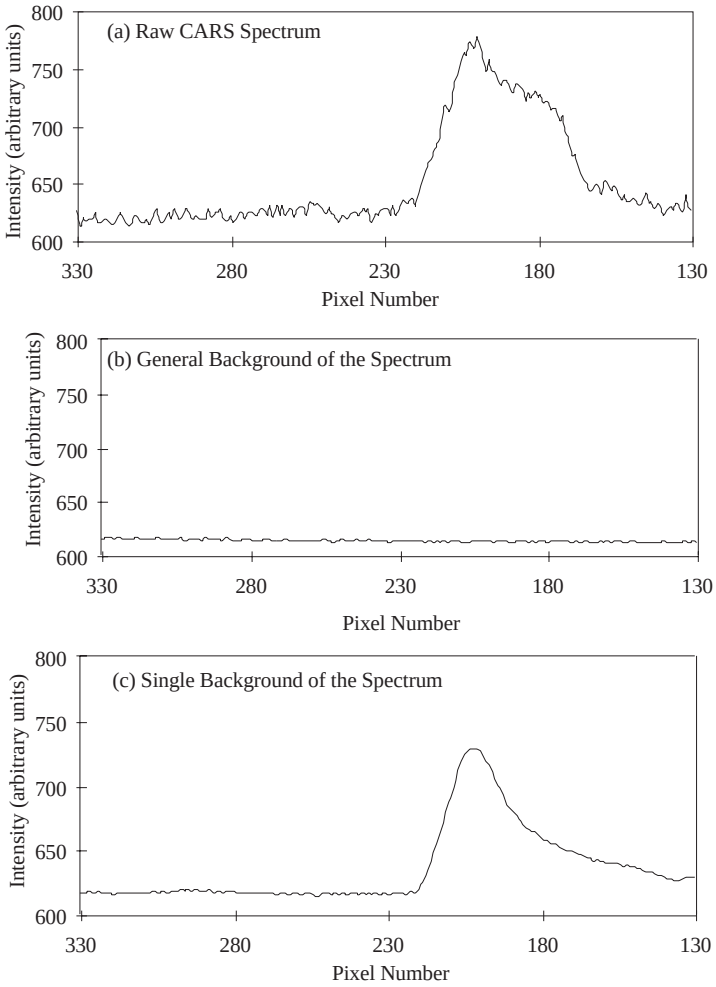

Figure 10. An unprocessed CARS Spectrum with general and single backgrounds (CH_4-air flame, P = 30.4 kPa, $\phi = 1.3$).

An indication of the "closeness of fit" between the experimental and theoretical spectra was used to obtain temperatures (Bradley *et al.*, 1998). In this criterion, referred to as the "Signal to Noise Ratio (SNR)", the parameter SNR was calculated using the following expression:

$$SNR = \sum_{i=1}^{N} (I_e - b)/N \Bigg/ \sqrt{\sum_{i=1}^{N} (I_t - I_e)_i^2/N} \quad (1)$$

where I_t is the intensity of the theoretical signal at wave-number, i, cm^{-1}, I_e is the intensity of the experimental signal at the same wave-number, b is the mean theoretical matched baseline intensity and N is the number of discrete wave-numbers over which the spectrum was measured. A 'noisy' spectrum is associated with a low SNR, and good spectrum is associated with a high SNR.

In the present study, the "closeness of fit" was determined by a combination of the SNR and visual examination. It was found that, in most cases, the SNR was effective in rejecting bad spectra and accepting good spectra. However, in approximately 5% of spectra, the SNR criterion produced unreliable results in the coal-air-methane flames. Therefore, in the present work, all spectra were visually inspected to improve reliability.

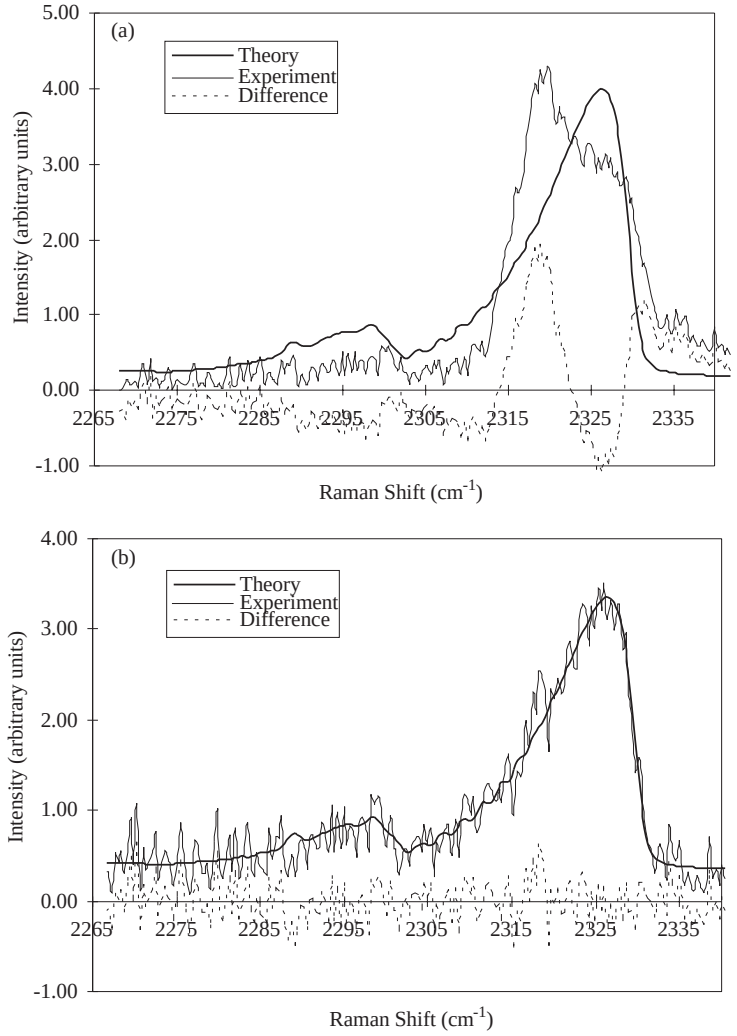

Figure 11. Theoretical fit of the CARS spectrum in Fig. 10a using (a) General background in Fig. 10b, and (b) Single background in Fig. 10c (CH_4-air flame, P = 30.4 kPa, $\phi = 1.3$).

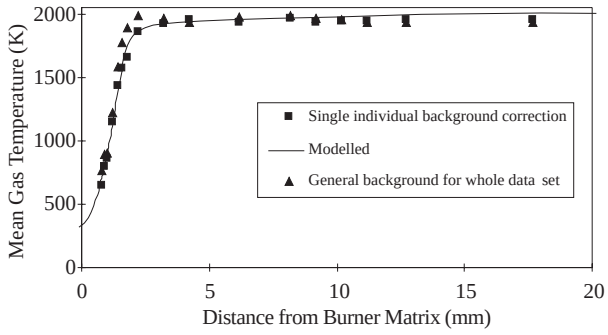

Figure 12. Temperature measurements and modelled predictions (methane-air flame, P = 30.4 kPa, $\phi = 1.3$)

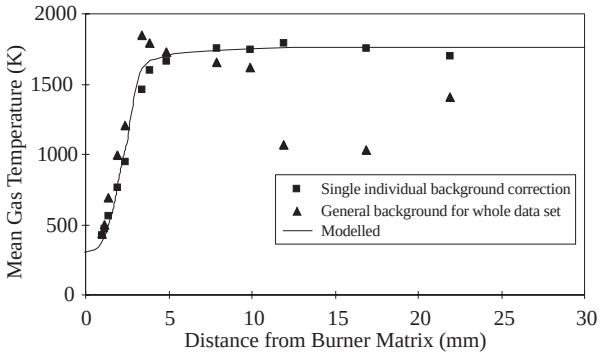

Figure 13. Temperature measurements and modelled predictions (coal-methane-air flame, P = 30.4 kPa, $\phi_{ov} = 2.276$, $\phi_{gas} = 1.273$)

Discussion and Conclusions

Methane-air flames

The CARS technique has been applied very successfully for clean methane-air flames ($\phi = 0.6$, 0.7 and 1.0) at 30.4 kPa. The measured temperatures, averaged over approximately 200 spectra, at each location are in good agreement with those computed from a one-dimensional mathematical model (Usta, 1999; Bradley *et al.*, 2001). The rms temperature variations arise from the inherent nature of the CARS technique. They increase in the reaction zone, as a consequence of the high temperature gradients, and are up to 13-25% of the mean gas temperature. The rms values subsequently decrease to approximately 3.5% in the post-flame zone, which is normal for this technique. Because the methane-air flames were 'clean', the minimum SNR in the present work was about 1.5, and most of the spectra were fitted very well by the theoretical spectra. More than 90% of all the CARS spectra recorded in these flames were suitable for full processing. The remainder were rejected because of excessive noise, indicated by excessive differences between the experimental and theoretical spectra.

Although the methane-air flame at $\phi = 1.3$ did not produce any soot, some C_2 emission was observed and this was independent of laser power, indicating that it was not laser induced. C_2 emission intensity was a function of position within the flame. Hence, the C_2 emission at every position was recorded and this background spectrum was subtracted from the CARS spectrum before attempting to fit it with a theoretical one. This technique was validated in a well characterised methane-air flame with $\phi = 1.3$. The rich methane-air flame measurements had a similar number of accepted spectra, SNR and experimental scatter, (represented by the rms temperature variation) to those in lean and stoichiometric flames, which did not suffer from the C_2 emission contamination of the CARS spectra. The experimental temperature distribution was in fairly good agreement with the theoretical results as were those in lean and stoichiometric flames.

Coal-air-CH_4 flames

Although the intensity of C_2 emission was independent of the presence of the laser beams in the methane-air flame ($\phi = 1.3$), this was not the case for coal flames. Moreover, its intensity depended upon the laser beam intensity, and persisted further downstream than does naturally occurring C_2 emission. Therefore, in coal flames it was necessary to measure background C_2 emission spectra in the presence of laser beams, but without CARS generation.

The range of measurements reported here covered seven overall equivalence ratios and two particle sizes. Sometimes particle-induced breakdown rendered a CARS spectrum unreliable. In addition, the signal strength sometimes was reduced excessively by attenuation of the laser beams in the particle-laden gases, although this was countered by the low particle number density as a consequence of the low pressure. Due to particles and particle breakdown in coal flames, excessive noise caused some "bad" spectra. Therefore, prior to acceptance, all spectra were examined and only those with fits between theory and experiment that yielded a SNR of over 0.6 were considered to be good. The proportion of useable spectra was reduced from a value of 90% in the methane-air flames to between 20 and 80 % by these effects and by the increased C_2 emission in coal-methane-air

flames. Although rms values increased in the reaction zone, where the temperature gradients are high, they decrease to approximately 5-10% in the post-flame zones. Although these values are higher than for CH_4-air flames, they are typical of coal-air flames reported elsewhere (Hancock *et al.*, 1992).

Nomenclature

b	mean theoretical matched baseline intensity
f	focus
I	signal intensity
i	wave-number
N	number of discrete wave-numbers
P	pressure

Greek symbols

ϕ	equivalence ratio
ν	frequency of radiation

Abbreviations

CCD	charge coupled device
SG	Gaussian function
SNR	signal to noise ratio
SL	Lorentzian function

Subscripts and superscripts

e	experimental
ov	overall
gas	gas
t	theoretical

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