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Dual-broadband CARS temperature measurements in hydrogen-oxygen atmospheric pressure flames

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Abstract. Dual-broadband coherent anti-Stokes Raman scattering (CARS) spectroscopy of the Q_{01}-branch of H_2 has been employed for thermometry in an atmospheric-pressure hydrogen-oxygen flame. The aim was to investigate the applicability of the technique for single-shot temperature evaluation and to analyse the precision of the measurements. The results are presented of temperature and relative H_2 density mapping of the flame in the temperature range of 700–2800 K. The achieved precision of single-shot measurements was 3%–5%.

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Temperature measurements in hydrogen-oxygen combustion by means of nonlinear optical techniques are still a problem that has not been solved completely. Of special interest is a question of the precision achievable as a result of single-laser-shot (10 ns pulse duration) measurements as they can provide investigations of turbulence fields existing in real-scale technical combustors.

The serious problem of combustion thermometry by coherent anti-Stokes Raman scattering (CARS) is to find the compromise between the sensitivity, the signal-to-noise ratio, the time of data acquisition, and the precision of temperature measurements. Due to the nonlinear and coherent nature of CARS process, beatings of pump lasers' frequency components lead to strong fluctuations in the intensity distribution of a CARS spectrum which in turn result in errors in temperature determination. The only way to overcome this difficulty is to perform some sort of averaging: either over time – that means to perform accumulations of consecutive CARS signals, or spectral averaging. The latter approach allows one not to lose available temporal resolution and to achieve reasonable precision in single-shot temperature measurements. There exist two ways to perform spectral averaging: one is to average the measured parameters over the molecular transitions in a broad frequency range (broadband CARS); the other – to use many spectral components of two broadband pump lasers for averaging over the spectrum of the driving force, exciting Raman-active molecular transitions. This approach, first proposed for vibrational CARS [1] and later on developed for rotational CARS [2–4], is known as the dual-broadband CARS (DBB-CARS) technique.

In most of the previous flame temperature measurements by CARS nitrogen was used as a probe gas. Broadband CARS spectroscopy of hot N_2 molecules makes it possible to reach a good precision in temperature measurements due to the significant number of rotational components in the observed Q-branches and efficient averaging over the spectra of molecular transitions (see, for example [5–7]). In hydrogen-oxygen combustion hydrogen may be used to measure temperature [8, 9]. However, spectral averaging over the transitions is not so efficient for broadband measurements using H_2, because only a few narrow lines are observed in a CARS spectrum even at temperatures as high as 2000–3000 K, which greatly limits attainable precision of temperature determination. Of course, only fuel-rich regions of a flame, where H_2 is present, can be investigated in this way. This is typically the case in rocket propulsion, where the engines always are run with an excess of H_2.

This paper is devoted to the employment of dual-broadband CARS for express temperature measurements (during one laser shot) in hydrogen-oxygen combustion. The aim of the present study was to estimate the attainable precision and spatial resolution of single-shot temperature measurements for DBB-CARS and to envision the applicability field of the technique to hydrogen-oxygen technical combustion diagnostics. It should be noted that in case of hydrogen thermometry DBB-CARS, especially in conjunction with modeless lasers, which eliminate the noise due to mode competition, seems to be preferable over the conventional broadband CARS [7–11], since the most efficient averaging over the spectra of laser radiation can be achieved, providing the improvement of the precision of temperature measurements.

1 Experimental

The laser part of the CARS spectrometer consisted of a Quanta Ray DCR-3D Nd:YAG laser delivering 150 mJ in

the second-harmonic beam and two broadband oscillator-amplifier dye lasers. Part of the 532-nm beam (≈ 30 mJ) was used as a CARS pump, the rest being used to pump the dye-laser system. The pump beam had 0.7 cm^{-1} bandwidth with mode spacings of 0.005 cm^{-1}.

Dual-broadband pumping was realized with the help of two oscillator-amplifier dye lasers. Dye laser I was a modeless Rhodamine 6G amplified spontaneous emission (ASE) dye laser operating around 553 nm with a bandwidth of $\approx 170\,cm^{-1}$ and output energy of about 3 mJ. Dye laser II was a Pyridine 1/DMSO laser with a glass substrate used as an output coupler of the oscillator. The dye laser II output power reached 1.5 mJ, its lasing being centered at approximately 713 nm with a bandwidth $\approx 350\,cm^{-1}$. The difference between the central frequencies of the two dye lasers ($\approx 4000\,cm^{-1}$) corresponded to the falling edge of the fundamental Q_{01}-branch of hot hydrogen, with the rotational lines Q(5)–Q(11).

The pump beams, adjusted in diameters and divergencies, with their pulses overlapped in time, were combined in a USED-CARS [12] geometry and focused into the probe volume with a 300-mm-focal-length lens. The beam spot of the pump laser, with the unstable resonator installed to provide the donut intensity profile, had a ring shape at the focusing lens, with the outer and inner diameters of 20 and 10 mm, respectively. The dye lasers' beam diameters were about 8 mm. Under these conditions the beam waist diameters in the focal region were estimated to be about 40 μm. The longitudinal spatial resolution for the employed interaction geometry, defined as the length of the volume in which over 90% of the CARS signal is generated, was measured to be 2.5 mm.

The CARS beam, after recollimation and preliminary filtering by dichroic and notch filters, was dispersed by a Jobin-Yvon HR1000 monochromator equipped with a 2400-grooves/mm holographic grating and was registered by a diode-array detector with a gated MCP image intensifier (Spectroscopy Instruments, IRY 1024). The width of the apparatus function of the monochromator was about 3 cm^{-1}, and that determined the observed linewidth of single rotational components of the H_2 Q_{01}-branch spectra, which were being detected in all the experiments.

Calibration measurements have been performed using stationary and well-controlled atmospheric pressure premixed hydrogen/air flame in a McKenna burner, exactly the same as employed in [13]. This burner has been recommended in [13] as a calibration device for flame temperature diagnostics techniques. Further measurements have been performed in a H_2/O_2 flame produced by an atmospheric-pressure burner with coaxial gas injection and turbulent mixing of the components. The burner had a central tube of 2.5 mm inner diameter, for supply of O_2, and of 4 mm outer diameter. The concentric slit going around this tube, for H_2 injection, was 0.25 mm wide. Gas flow rates were measured by electronic flow controllers (Tylan), preliminarily calibrated by direct measurements. The operation of the controllers is based on the effect of heat conduction.

The main principle of this kind of injectors is that O_2 and H_2 leave the burner plate separately, in concentric turbulent jets. Vortices generated in the shear layer between these jets, as well as the diffusion process, provide mixing of the components. Characteristic features of the investigated flame are high gradients of H_2 concentration and clearly observable mixing zones. This is the reason that CARS beam intensity from different parts of the flame varies several orders of magnitude. In most of the measurements H_2 flow was maintained at 13.2 slpm and O_2 flow at 3.8 slpm, respectively, so that the mass equivalence ratio was 1.7. At these conditions the slightly visible flame torch was about 150 mm high and 20 mm in diameter in its widest part.

In order to vary the location of the probe volume inside the axisymmetric flame under study, the burner was mounted on an X-Z translation stage (with Z axis coincident with the axis of gas injection, and X axis pointing in the transverse direction). CARS spectra have been recorded in several flame cross sections, with the step of 100 or 200 μm in X direction, from the height of $Z = 3$ mm to $Z = 92$ mm above the burner surface. The probe volume was oriented along the Y axis, its centre being positioned in the XZ plane.

2 Results and discussion

The main aim of temperature measurements in the McKenna burner using DBB CARS of H_2 Q-branch, in the range of 1400–1900 K, was to verify the employed experimental technique and data processing routines and to estimate the precision of measurements. So, in order to facilitate the direct comparison of our results with those obtained by other authors applying usual broadband N_2 CARS thermometry, the experimental conditions have been chosen to coincide exactly with those used in [13]. This means that all the temperature measurements have been performed 15 mm above the centre of the burner surface, while hydrogen flow rates and mixture stoichiometries Φ were adjusted to be the same as in [13].

Some comments should be made about the data processing approach. At the beginning and at the end of each series of data acquisition (in 10–15 min) accumulated nonresonant spectra from room air have been recorded and later on employed for normalization of the obtained H_2 Q-branch spectra. As temperatures T in the premixed flame were rather high (1400–1900 K) and gas densities were quite low ($\approx$ 0.15–0.2 Amagat), the Q-branch lines were considered to be inhomogeneously Doppler-broadened and having practically the same widths, with negligible manifestations of both the Dicke narrowing and homogeneous J-dependent collisional broadening by surrounding molecules, primarily N_2 and H_2O [14, 15]. Thus, under conditions of our experiments, a spectrally integrated intensity (line strength) I_J in a broadband CARS spectrum, after subtraction of the nonresonant background pedestal (if present), can be to a good approximation regarded as proportional to the square of the population N_J of the lower rotational level of the transition and independent on the transition linewidth. To the contrary, the weak J-dependence of the Raman cross section for a given vibrational band of H_2 molecules and of the Stokes laser frequency factor in the line strength should be taken into account, in accordance with [16]. Thus, temperatures were derived for each spectrum from the standard least-squares linear fit for the populations of the rotational levels of the $v = 0$ state versus the energies E_J of these levels, in accordance with the Boltzmann formulae. Only the strong odd-J Q-branch components of the $v = 0 \rightarrow v = 1$ band ($Q_3 - Q_9$) were regarded.

The lines of the $v = 1 \rightarrow v = 2$ hot band were not processed, even if observed.

The random shot-to-shot errors in temperature values determined from a series of single-shot spectra appear as a result of the normalization procedure, as the accumulated nonresonant CARS spectra were employed for this purpose. It should be also noted, that the simple approach used presently for temperature determination may produce systematic errors, which in our case of high temperatures and low densities appear mainly because of the uncertainties in determination of the nonresonant background in CARS spectra. Application of more sophisticated methods of data treatment, that employ the simulation of the shape of experimental Q-branch spectra, including the observable weak even-J and hot band lines, is expected to increase the accuracy of temperature determination.

The results of the experiments in the McKenna burner are summarized in Table 1. Column 3 contains the results of temperature measurements performed in [13] (the errors have been recalculated on the basis of the stated 2.5% accuracy), whereas columns 4, 5, and 6 contain our results. Temperature values T_A in column 4 are derived, for comparison with [13], by processing a single spectrum which was accumulated on the multichannel detector during a given number of consecutive laser shots. Values in columns 5 and 6 are the results of a fit of temperature histograms, obtained with the bin width equal to 50 K from 300 single-shot spectra, to a Gaussian probability distribution function, centred at T_H and with a standard deviation σ. The relative standard deviation of temperature (in %) is given in brackets in column 6.

The results presented in Table 1 lead to the following conclusions.

(i) All our temperature measurements provided by processing of the accumulated spectra (T_A) coincide, within the experimental errors, with those presented in [13].

(ii) The temperatures T_A are systematically (by $\approx 2\%$) lower than those derived from the histogram analysis (T_H). This fact can be mainly attributed to the character of the accumulation procedure, when spectral lines, more intensive at lower temperatures, give more significant contributions to the derived temperature values. It should be noted, in addition, that single-shot spectra can not be normalized in a proper way during the accumulation on the detector.

(iii) The results of processing of different histograms, obtained at the same experimental conditions in independent series of measurements, are well reproducible and coincide with each other within the errors quoted in columns 5 and 6. Thus, estimated precision of the DBB-CARS thermometry at 1400–1900 K was below 0.2% for determination of temperature as a histogram center T_H, being an average over 300 shots (see Fig. 1), and in the range of 3%–5% for single-shot measurements. Hence, if we take the values from [13] as the calibration ones, the accuracy of single-shot measurements with our experimental setup can be estimated to be at the level of 5%–6%.

Further experiments have been aimed at performing temperature mapping of the axisymmetric flame of the atmospheric pressure H_2/O_2 turbulent burner described above. Again, two types of measurements have been performed, and either accumulated spectra or single-shot histograms (300 shots in each series) were analysed. The spectra with signal-to-noise ratio worse than 10 were neglected, thus precluding the measurements above $Z = 95$ mm height.

The accumulated spectra were used to determine temperature as T_A, the range of measured values being 700–2800 K. The number of accumulations varied from 10 to 500, depending on spectra signal-to-noise ratio. An example of an accumulated spectrum is presented in Fig. 2. The procedure, similar to the one described above for the case of the McKenna burner, was used to derive temperature. As previously, only up to 4 odd-J lines ($Q_3 - Q_9$) were used in the Boltzmann plot of the rotational level populations. Within the experimental errors two different slopes were not observable in this plot, and CARS spectra obtained, both accumulated and single-shot ones, were described by a single temperature.

If T is known, the spatial distribution of H_2 number density N can be estimated from the line strength I_J of

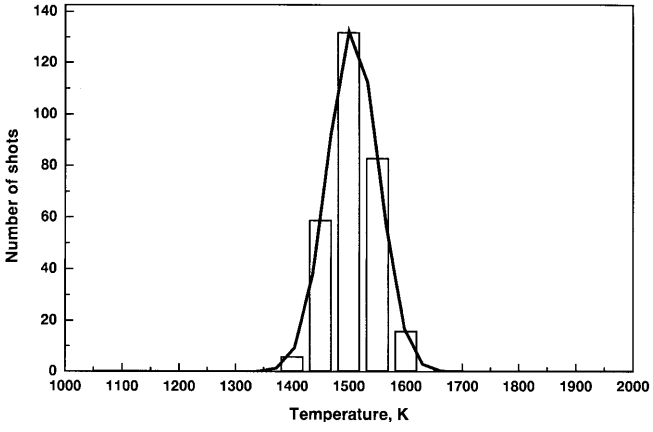

Fig. 1. Typical temperature histogram obtained in the McKenna burner at H_2 flow rate of 19.7 slpm and $\Phi = 1.4$. The *solid line* corresponds to the Gaussian distribution with $T_H = 1507$ K and $\sigma = 44$ K

Table 1. Results of the experiments in the McKenna burner

Experimental conditions		Derived temperature /K [13]	Derived temperature data /K Present work		
1	2	3	4	5	6
H_2 flow rate (slpm)	Stoichiometry, Φ	300 accumulated broadband N_2 CARS spectra	Accumulated spectra, T_A	Histogram center, T_H	Histogram halfwidth, σ
9.85	1.2	1452 ± 36	1536 ± 90	1540 ± 3	80 ± 2.5 (5.2)
9.85	1.3	1374 ± 34	1398 ± 40	1430 ± 1	53 ± 0.5 (3.7)
19.7	1.2	1641 ± 41	1694 ± 110	1705 ± 2	92 ± 2.5 (5.4)
19.7	1.3	1558 ± 39	1552 ± 100	1590 ± 2	56 ± 1.5 (3.5)
19.7	1.4	1503 ± 38	1463 ± 90	1507 ± 1	44 ± 0.15 (2.9)
34.6	1.2	1873 ± 47	1888 ± 150	1913 ± 2	99 ± 2.0 (5.2)
34.6	1.3	1775 ± 44	1738 ± 130	1770 ± 1	66 ± 1.5 (3.7)

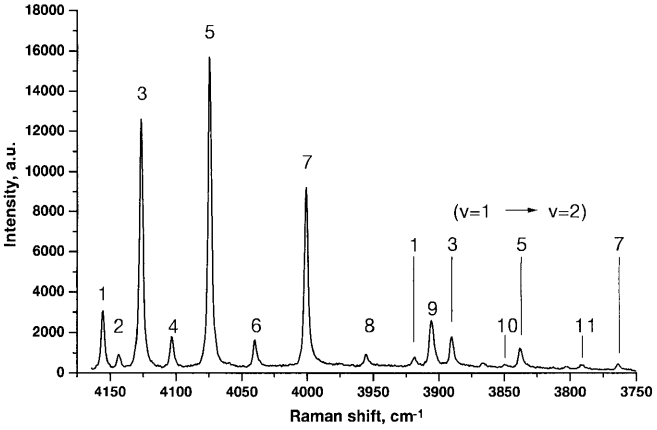

Fig. 2. DBB-CARS spectrum of H_2 from a burner with a coaxial hydrogen-oxygen flame (CHOF) at $Z = 21$ mm, $X = 3$ mm, and $\Phi = 1.7$. Number of accumulations -10, $T_A = 2880 \pm 150$ K

a Q-branch component of a CARS spectrum using, for the case of Doppler-broadening, the relation:

$$N(T) \approx \frac{Z_R(T)(1 + e^{-E_v/kT})}{1 - e^{-E_v/kT}} e^{E_J/kT} (I_J)^{1/2}, \quad (1)$$

where $Z_R(T)$ is the rotational partition function for H_2, $E_v = 4161\,\mathrm{cm}^{-1}$ is the vibrational energy in the $v = 1$ state, E_J is the energy of a rotational level with a given J.

Figure 3 presents examples of radial distributions of temperature and of H_2 number densities measured in the flame with the mass equivalence ratio of 1.7 at different heights above the burner surface. These results allow the characteristic features of the combustion process in different regions to be distinguished.

The distribution of H_2 density measured at $Z = 3$ mm (Fig. 3a) is characterized by significant gradients and 1 mm width (at half height) which is indicative of weak turbulent and diffusion penetration of H_2 towards the stream axis. Temperature distribution is characterized by relatively low values ≈ 1000 K in the vicinity of the stream axis, inside the O_2 jet, which also points to weak mixing of the concentric jets. Significant temperature increase in the peripheral part of the flow (at $X > 4$ mm) is due to turbulent and convective mixing of H_2 with room air, which maintains efficient combustion with a high heat release in this region. As expected, the lowest temperatures are observed near the centre of the H_2 jet, located at the position of the injection slit ($X = 2.125$ mm), where H_2 densities are the highest and where neither O_2 nor room air are able to penetrate.

Up to the height of $Z = 15$ mm above the burner surface H_2 and O_2 mix up efficiently and temperatures reach 2600–2800 K at $X = 0$–3 mm (Fig. 3b). The distribution of H_2 density is characterized by a larger width of about 5 mm, its center being displaced from the burner axis to $X \approx 4$ mm. In the peripheral parts of the flame the combustion demonstrates the same features as at lower heights above the burner surface, as far as H_2 has not been burnt out, and combustion is still maintained by mixing of H_2 with room air. The temperature minimum is still observed at the position of the peak of H_2 density, revealing the presence of the significant excess of heated H_2 in the form of a residual jet.

At the flame heights above $Z = 50$ mm radial temperature distributions are nearly flat and the measured temperature values are in the range of 2800 K, gradually decreasing towards the periphery of the flame at $X > 10$ mm. This may be regarded as an indication that H_2 and O_2 have been completely mixed and the injected H_2 has been practically burnt out. The density profiles of the residual H_2 also become almost uniform.

Thus, the measured temperature distributions at different heights show that in the process of turbulent mixing and combustion under study, the mixture components start to react being preheated up to 1000–2000 K. For comparison, the results should be mentioned of the theoretical calculations, in the frame of the model described in [17], of adiabatic combustion for a stoichiometric H_2/O_2 mixture. If the mixture is ignited at 293 K the calculated final temperature of the combustion products should be 3050 K. The enhancement of the initial temperature of mixture components by 1000 K increases the final temperature only by ≈ 100 K. Hence, one should expect that under our experimental conditions, though far from the adiabatic ones, the observed preheating of the mixture components in course of the turbulent combustion will probably also not result in significant enhancement of the flame temperature.

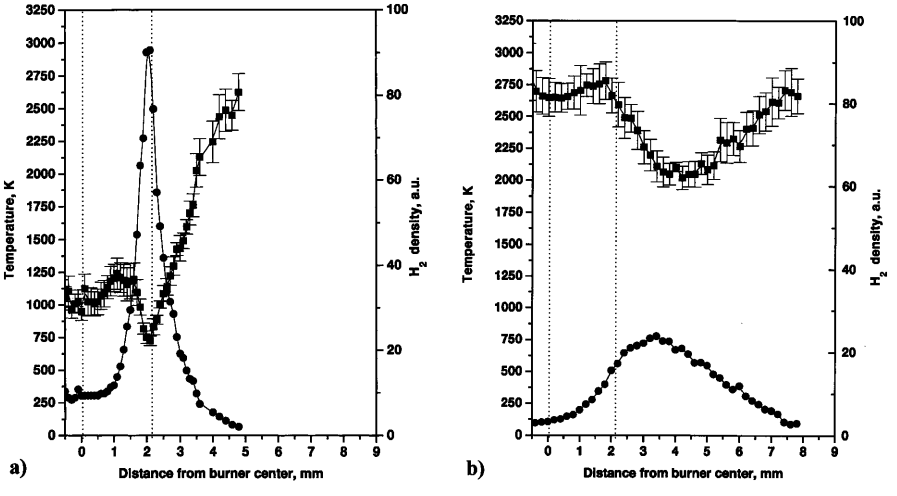

Fig. 3a,b. Radial temperature (*squares*) and H_2 density (*circles*) distributions in the CHOF: **a** $Z = 3$ mm, **b** $Z = 15$ mm. The *dotted lines* show the positions of the O_2 jet axis ($X = 0$ mm) and of the H_2 injection slit ($X = 2.125$ mm)

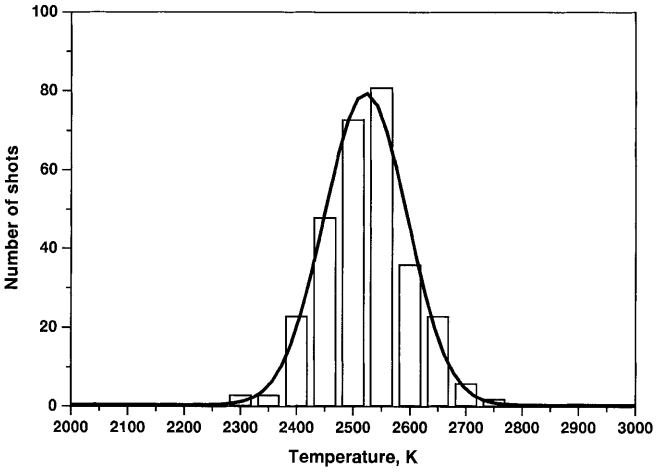

Fig. 4. An example of a temperature histogram obtained in the CHOF: $P = 1$ bar, $Z = 15$ mm, $X = 3$ mm, $\Phi = 1.7$. The *solid line* corresponds to the Gaussian distribution function with $T_H = 2522$ K and $\sigma = 73$ K

The analysis of the series of single-shot temperature measurements in the turbulent burner shows that typically the widths of the obtained shot-to-shot temperature histograms are not broader than those measured for the McKenna burner. One of the temperature histograms for the turbulent flame is presented in Fig. 4. It shows that the dispersion of temperature does not exceed 3% at ≈ 2500 K. This leads to a conclusion that in the investigated flame regions and at the combustion parameters, typical for our measurements, the fluctuations of local temperatures in the probe volume are relatively small.

3 Conclusion

The results obtained show that DBB-CARS spectroscopy of hydrogen-oxygen flames is applicable to temperature measurements in the broad temperature range of 700–2800 K and allows us to realize single-shot measurements with the precision of about 3%–5%, assuming comparatively high signal-to-noise ratios are provided. This temperature range and precision are of interest for diagnostics of high-pressure turbulent flames. Performed qualitative analysis of the measured radial temperature and concentration distributions allows us to observe the characteristic features of hydrogen-oxygen combustion in the atmospheric-pressure turbulent burner with the coaxial gas injection.

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