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High-resolution line-shape spectroscopy during a laser pulse based on Dual-Broad-Band – CARS interferometry

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Abstract. **A high-resolution spectroscopic method is developed for recording Raman spectra of molecular transitions in transient objects during a laser pulse with a resolution of ~ 0.1 cm^{-1}. The method is based on CARS spectroscopy using a Fabry – Perot interferometer for spectral analysis of the CARS signal and detecting a circular interferometric pattern on a two-dimensional multichannel photodetector. It is shown that the use of the Dual-Broad-Band – CARS configuration to obtain the CARS process provides the efficient averaging of the spectral – amplitude noise of the CARS signal generated by a laser pulse and, in combination with the angular integration of the two-dimensional interference pattern, considerably improves the quality of interferograms. The method was tested upon diagnostics of the transient oxygen – hydrogen flame where information on the shapes of spectral lines of the Q-branch of hydrogen molecules required for measuring temperature was simultaneously obtained and used.**

Keywords: *CARS, collision broadening coefficient, Fabry – Perot interferometry, combustion thermometry, hydrogen – oxygen combustion.*

1. Introduction

The use of CARS spectroscopy for thermometry of oxygen – hydrogen combustion at high pressures and high temperatures based on the recording of spectra of the Q-branch of hydrogen [1 – 4] has shown that for correct measurements of the gas temperature, it is necessary to know the coefficients of collision broadening of spectral lines of the Q-branch by H_2O molecules along with their temperature dependence and the dependence of the linewidth or the collision broadening coefficient on the rotational quantum number J.

The coefficients of collision broadening of the spectral lines of the hydrogen Q-branch by N_2, H_2, and H_2O molecules were earlier measured at temperatures below 1800 K in papers [5 – 7]. A gas mixture in a heated high-pressure cell filled with hydrogen and water vapour was studied by the method of high-resolution stimulated Raman amplification of light with a cw single-frequency tunable pump source. As pointed out in [5, 6], the main factor restricting an increase in the temperature of the gas mixture in the cell was a high chemical activity of water vapour at high temperatures under stationary conditions.

In [4], we proposed the method for measuring the width of lines of the Q-branch of hydrogen by CARS spectroscopy directly in an oxygen – hydrogen combustion chamber operating in the regime of repetitively pulsed replacement of the mixture at a high temperature ($T \sim 2000 - 3000$ K) and pressure ~ 100 bar. We measured the collision broadening coefficients of different lines of the Q-branch of hydrogen broadened in collisions with water molecules. Note that the collision broadening coefficients of the Q_1, Q_3, Q_5, and Q_7 lines of hydrogen coincide within the experimental error with the values extrapolated from the values of broadening coefficients measured at 1800 K [5 – 7].

While the experimental and extrapolated broadening of hydrogen lines can be successfully used together with model calculations for the CARS thermometry of oxygen – hydrogen flames in the temperature range 300 – 2700 K [4 – 7], the problem of correct temperature measurements in oxygen – carbon flames still remains unsolved. This is mainly explained by insufficient information on the broadening coefficients for hydrogen lines in collisions with N_2, CO_2, etc. at high temperatures. In addition, experimental problems appear because it is necessary to measure simultaneously the concentrations of H_2O, CO_2, and N_2 in combustion products during a single laser pulse. The accuracy of temperature measurements from CARS spectra is critical to the reliability of the values of line broadening coefficients and their temperature dependence, and to the measurement accuracy of concentrations of colliding particles. Thus, the development of methodical aspects of high-temperature thermometry and related high-resolution line-shape CARS spectroscopy during a laser pulse still remains of current interest.

2. Experimental

We developed earlier [4] the high-resolution spectroscopic method for recording the CARS spectra of molecular transitions of the Q-branch of hydrogen in transient media during one laser pulse with a resolution of ~ 0.1 cm^{-1}. The method uses a Fabry – Perot interferometer for spectral

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analysis of the shape of Raman lines by recording the interference pattern with a linear CCD array. The CARS spectrum of the transition of the Q-branch is recorded during one laser pulse by using broadband (BB) CARS. We measured by this method the collision broadening coefficients for a number of lines of the Q-branch; however, the measurement error of these coefficients was as high as $\sim 30\,\%$.

The aim of this paper was to reduce the error of linewidth measurements by using the CARS schemes allowing a more efficient averaging of the CARS signal noise during one 10-ns laser pulse.

Our studies have shown that the quality of interference patterns is deteriorated mainly due to the spectral noise of a dye laser. Moreover, we found that, irrespective of the dye laser design (with a resonator containing various selecting elements or without a resonator, operating in the amplified spontaneous emission regime), the spectral profile of a CARS signal almost always had a distinct noise structure within the spectral width of one vibrational–rotational transition (see Fig. 2).

To solve this problem and improve the quality of the line profile obtained for one laser pulse, we used the CARS scheme with two broadband pump lasers (Dual-Broad-Band (DBB) CARS [8, 9]). This approach reduces the spectral noise of a CARS signal (in our case, it is generated within the contour of one line) obtained for one laser pulse. This occurs due to the averaging of the spectrum of a driving force (determined by the convolution of the spectra of two broadband pump lasers) during the interaction of these lasers with molecules of the gas under study in the case of a Raman resonance.

The additional noise in the intensity profile of interferograms is caused by speckles observed in the CCD detector plane due to the use of a multimode optical fibre for the transport of a CARS signal. To reduce the line profile distortions introduced by this noise, we performed angular averaging over the circle of circular Fabry–Perot interferograms recorded with a two-dimensional CCD array.

The optical scheme of the experimental setup is presented in Fig. 1. We used two dye lasers (yellow and red) with relatively broad linewidths ($\sim 5\ \mathrm{cm}^{-1}$) in the DBB–CARS scheme. On the one hand, the linewidths of the lasers were small enough for their convolution to have the width smaller than the distance between the lines of the Q-branch ($20-100\ \mathrm{cm}^{-1}$), and on the other, these widths are sufficient for efficient averaging over the emission spectra of dye lasers. The difference of the central frequencies of emission spectra of these lasers was tuned to the frequency of the transition being probed. The second harmonic of a single-frequency (linewidth ~ 100 MHz) pulsed Infinity Nd^{3+} : YAG laser (Coherent Radiation) was used as the third pump wave for CARS. The typical output pulse energy of this laser at 532 nm was 100–200 mJ for 3-ns pulses.

A part of the output energy ($\sim 10-15$ mJ) was used to pump CARS and the rest of the energy was used to pump both dye lasers. In the red dye laser the solution of pyridine dye in DMSO was used, and the output pulse energy was 3 mJ. The yellow dye laser operated on the solution of Rhodamine 6G in methanol and produced 2 mJ of output pulse energy. The emission frequency of the red laser could be varied to tune the difference of the central frequencies of the two dye lasers ($\sim 4000\ \mathrm{cm}^{-1}$) to the combination frequencies of the $Q_1 - Q_9$ lines of the hydrogen molecule. All the three laser beams were focused either inside a pulsed high-pressure combustion chamber or into a cell containing a gas under stationary conditions. The CARS spectrum was generated per laser pulse with a pulse repetition rate of 1 Hz.

The Fabry–Perot interferometer (Fig. 1) used to analyse the CARS spectra had a free spectral range of $1.67\ \mathrm{cm}^{-1}$ and a finesse of 12–18. The specially developed software was used for reading the recorded interferograms, calculating their centre of symmetry, angular integrating, and determining the parameters of the spectral line profile.

3. Results and discussion

In our earlier BB–CARS experiments [4], pumping was performed by the second harmonic of a single-frequency Nd^{3+} : YAG laser (the pump wave) and the dye laser line of width $\sim 5\ \mathrm{cm}^{-1}$ (the Stokes wave). We found that, irrespective of the dye laser design (we used lasers with the resonator and different mode structures and a laser without the resonator), the spectral noise of laser radiation was considerable and irreproducible from pulse to pulse.

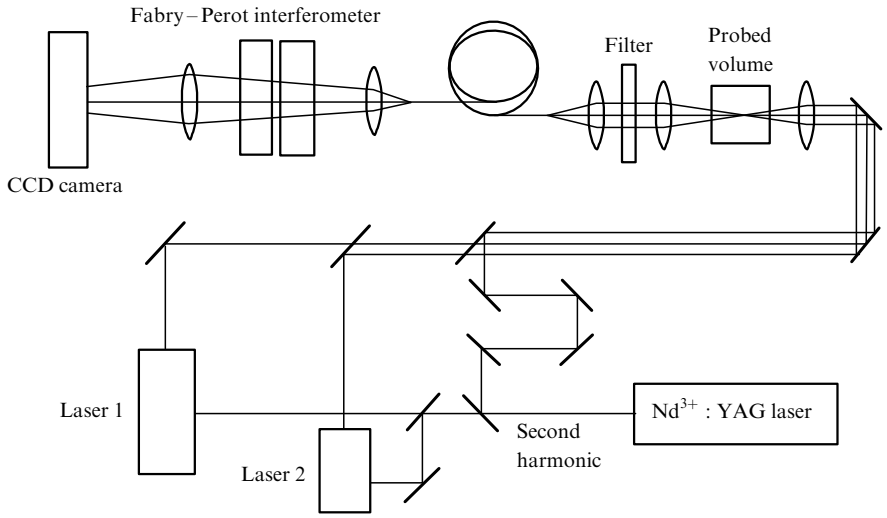

Figure 1. Optical scheme of the experimental setup.

This irregular noise was broadband and had Fourier components of width comparable with the width of Raman lines. The noise modulation gave rise to random spectral structures on the profile of lines in the CARS spectrum of the Q-branch, which can be observed in a series of interferograms presented in Fig. 2. Moreover, many interferograms were so noisy that it was impossible to use them to obtain information on the spectral line shape. Such interferograms were not considered, which in turn resulted in the increase in statistical errors in linewidth calculations.

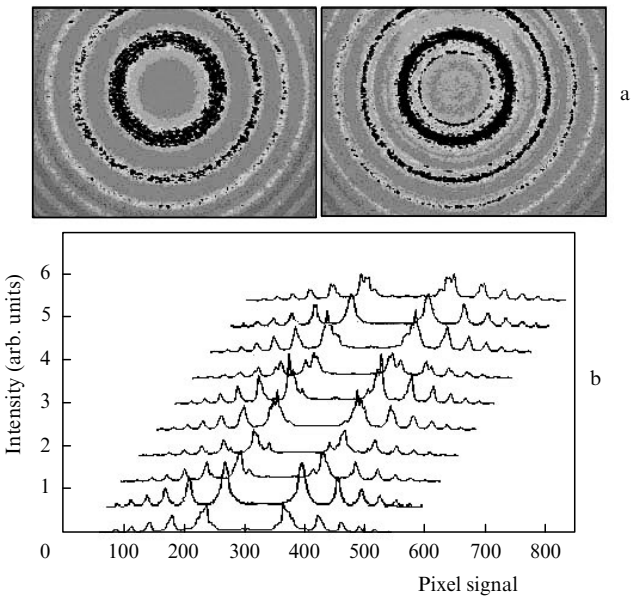

Figure 2. (a) Circular structure of Fabry–Perot interferograms for two successive laser pulses (BB–CARS configuration) obtained in a cell with the H_2-N_2 mixture (the H_2 and N_2 pressure is 3 and 42 bar, respectively) at room temperature (random speckle-modulation of the circular pattern is observed over the entire light field, as well as the fluctuating substructure of interference fringes caused by the spectral fluctuations of a Stokes laser from pulse to pulse) and (b) the results of angular integration of interferograms for a series of 10 laser pulses obtained in the BB–CARS configuration in a cell with a H_2-N_2 mixture (3 and 42 bar, respectively) at room temperature; the random intensity modulation of spectral lines caused by fluctuations of the spectrum of the Stokes laser from pulse to pulse are clearly observed; the further processing of the spectra requires selection.

Due to the use of the DBB–CARS method and the two-dimensional detection of the circular structure of Fabry–Perot interferograms followed by their angular integration, almost all interferograms obtained in our experiments during one laser pulse became suitable for linewidth measurements (Fig. 3).

To compare the BB–CARS and DBB–CARS methods, we measured the Q_1 linewidths under the same conditions in a cell with a mixture of hydrogen and nitrogen at pressures of 1 and 40 bar, respectively (Fig. 4). The instrumental function width in both cases was 0.09 cm^{-1}. Figure 4 shows the histograms of the Q_1 linewidth for hydrogen obtained by these methods. In both cases, the average linewidth of the transition was 0.16 cm^{-1}, the histogram width for the DBB–CARS and BB–CARS configurations was 0.006 and 0.017 cm^{-1}, respectively. We can conclude that the DBB–CARS method provides a better accuracy of linewidth measurement during one laser pulse.

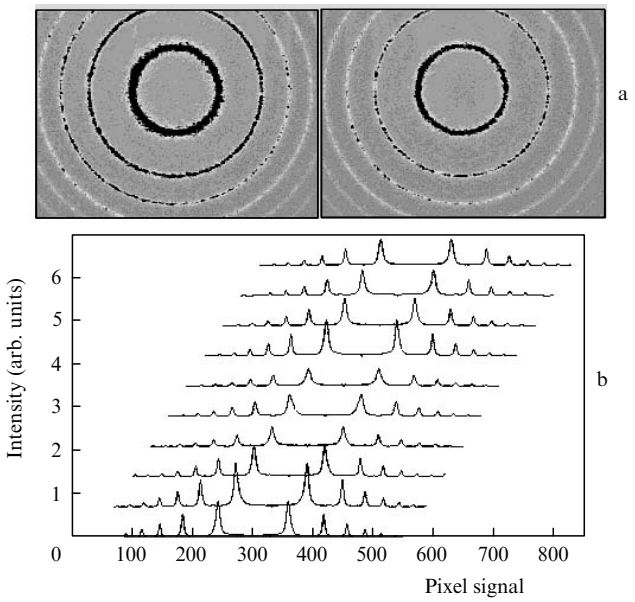

Figure 3. (a) Circular structure of Fabry–Perot interferograms for two successive laser pulses (DBB–CARS configuration) obtained in a cell with the H_2-N_2 mixture (the H_2 and N_2 pressure is 3 and 14 bar, respectively) at room temperature (only random speckle-modulation of the circular pattern is observed over the entire light field) and (b) the results of angular integration of interferograms for a series of 10 laser pulses obtained in the DBB–CARS configuration in a cell with the H_2-N_2 mixture (3 and 14 bar, respectively) at room temperature; all the spectral lines have good quality and are suitable for further processing.

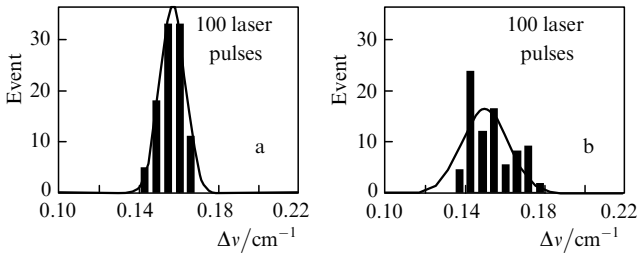

Figure 4. Histograms of the Q_1 linewidths in hydrogen measured by the DBB–CARS (a) and BB–CARS methods under identical experimental conditions in the H_2-N_2 mixture (the H_2 and N_2 pressure is 1 and 40 bar, respectively) at room temperature. The average linewidth is 0.16 cm^{-1}, the histogram width for the DBB–CARS and BB–CARS configurations is 0.006 and 0.017 cm^{-1}, respectively.

Based on the results obtained, we can propose a new method for measuring temperatures of flames with a hydrocarbon fuel during one laser pulse by using hydrogen as a probe molecule. The idea of the method consists in the simultaneous recording of the spectrum of the Q-branch of hydrogen with a grating spectrograph (line-intensity measurements) and an interferometer (linewidth measurements) used successively in one optical path and having orthogonal dispersions. A similar scheme was successfully used for measuring the density of a mixture of D_2 with Ar in a stationary cell [10]. In this method, information on the linewidth obtained from the DBB–CARS spectrum during one laser pulse is directly used for determining the relative populations of vibrational–rotational states and thereby temperature and does not require the knowledge of the concentration of components of the mixture, which cause

the broadening of spectral lines of probed molecules during collisions.

4. Conclusions

The use of the DBB–CARS configuration (averaging over the emission spectra of pump lasers) together with the angular integration of two-dimensional interferograms considerably improves the quality of interferograms obtained by measuring the widths of lines of the Q-branch of hydrogen during one laser pulse. As a result, almost all the DBB–CARS interferograms recorded in a repetitively pulsed high-pressure combustion chamber during one laser pulse became suitable for linewidth measurements. These results also suggest that this method can be used not only for the CARS thermometry of oxygen–hydrogen flames but also for studying flames in which carbon compounds are used as a fuel and the air oxygen is used as an oxidiser.

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