Appl. Phys. B 65, 619–624 (1997)

Applied Physics B Lasers and Optics



# The analysis of the precision of single shot 2λ-CARS temperature measurements in hydrogen

**W. Clauß[1], D.N. Kozlov[2], R.L. Pykhov[2], V.V. Smirnov[2], O.M. Stel'makh[2], K.A. Vereschagin[2]**

[1] German Aerospace Research Establishment (DLR), Institute of Chemical Propulsion and Engineering, Energy and Propulsion Section, D-74239 Lampoldshausen, Germany
[2] General Physics Institute of the Academy of Sciences of Russia, Vavilov Str. 38, 117942 Moscow, Russian Federation

Received: 16 October 1996/Revised version: 17 March 1997

**Abstract.** The results of the experiments on the precision of two-wavelength rotational CARS thermometry (2λ-CARS thermometry) of hydrogen are presented. The temperature was determined in a heatable sample cell in the range 300–1100 K and at pressures 1–25 bar was determined by using the measured values of single-shot and averaged CARS energies for the rotational transitions of the S branch. CARS beams were generated by using in turn three types of Stokes laser, with different spectral properties. A second cell, also filled with hydrogen, was established to obtain the reference signals. Relative standard deviations $\sim 1.5\%$ at 296 K and $\sim 8\%$ at 1000 K were achieved for temperature values measured in single laser shots. The precision of the measurements at different spectral characteristics of the pump lasers, linewidths of the probed Raman transitions, and referencing conditions, with variations in pressure and temperature in the sample volume, is analyzed and possibilities for its enhancement are discussed.

**PACS:** 33.20.Fb; 82.40.Py

Gas-temperature measurements based on rovibrational CARS (coherent anti-Stokes Raman scattering) spectroscopy are widely used in many applications [1]. Two aspects are the most important: the acquisition time in the recording of the spectra and the precision of the values obtained temperature. In general, amplitude and phase fluctuations in the laser fields lead to significant noise in the spectral shape and integral energy of CARS, so that conditions of spectral (over spectra of the lasers employed and molecular transitions) or temporal averaging should be provided to achieve the desirable precision of temperature evaluation. For temperature measurements under non-stationary conditions, temporal averaging is not applicable, and pulsed CARS employing at least one broadband pump laser to record a significant portion of the spectrum during one laser shot should be used. This provides efficient spectral averaging and good precision in the temperature measurements if a large number of transitions of the probe molecules are involved, as in the example of nitrogen [2–4].

With hydrogen as a probe molecule, only a few narrow lines are detectable in the CARS spectrum even at temperatures as high as 2000–3000 K [3, 5] and averaging over transitions is not so efficient for single-shot broadband CARS thermometry. In this case the two-wavelength CARS (2λ CARS), previously proposed in [6, 7], is preferable to the conventional broadband CARS. In 2λ CARS only two transitions are simultaneously excited by using two Stokes lasers and the pulse energies in each of the CARS beams are measured. Here, averaging over the spectra of laser radiation can provide the improvement in the precision of the temperature measurements.

Some problems in the application of CARS for thermometry of hydrogen were discussed in our previous paper [8], devoted to the development of $H_2$-based CARS temperature diagnostics of high-pressure hydrogen–oxygen combustors, characterized by strong temperature and density fluctuations. The present study continues the preliminary work reported in [8]. Its aim is to investigate the precision of single-shot rotational 2λ-CARS thermometry of $H_2$ under steady-state conditions in relation to the spectral characteristics of various Stokes lasers used in the CARS spectrometer, the linewidths of the probed Raman transitions, and the conditions of signal referencing at different pressures and temperatures in the sample volume. The transitions $S_1$ and $S_3$ of the rotational S-branch spectrum of hydrogen (with Raman shifts of 587 $cm^{-1}$ and 1035 $cm^{-1}$, respectively) were excited. Single-shot and averaged temperature measurements were performed in a heatable high-pressure cell with neat hydrogen at pressures up to 25 bar and temperatures up to 1100 K.

## 1 Experimental set-up

The scheme for the 2λ-CARS spectrometer employed in our experiments has been described in detail elsewhere [8]. Briefly, it consists of a repetitively pulsed Q-switched Nd: YAG pump laser with frequency doubling (10 Hz, bandwidth $\sim 0.7\ cm^{-1}$, mode spacing $\sim 0.005\ cm^{-1}$) and a dye laser module providing Stokes radiation at two wavelengths adjustable near $\sim 549$ nm and $\sim 563$ nm. Along with the narrowband ($\sim 0.3\,\mathrm{cm}^{-1}$) distributed feedback dye lasers (DFDL) used previously [8], two other configurations of the dye laser module were tested in a comparative study of the influence of spectral characteristics of the laser system on the CARS signal noise.

*(a) Amplified spontaneous emission dye lasers (ASEDL).* To provide modeless laser radiation a pair of longitudinally pumped dye oscillators was used, based on two-pass amplification of spontaneous emission, with a grating in the Littrow configuration [9] for narrowing of an ASE spectrum. This method of generation of modeless radiation is similar to that proposed in [10]. A slightly saturated amplifier stage was added to decrease fluctuations in the intensity. The spectral width of both the ASE lasers was $\sim 30$–$50\,\mathrm{cm}^{-1}$, with a pulse energy of $\sim 23\,\mathrm{mJ}$.

*(b) Broadband dye lasers.* Two broadband dye lasers together with the second harmonic of the Nd: YAG laser were used to realize dual-broadband, nondegenerate scheme of CARS [11] on $S_1$ transition. For each of the dye lasers a two-mirror resonator configuration was used. Both lasers had similar pulse energies of about 3 mJ. The total spectral width was $\sim 150$–$180\,\mathrm{cm}^{-1}$ with a mode spacing of $\sim 0.03\,\mathrm{cm}^{-1}$. The central wavelengths were shifted towards the maxima of the fluorescence of the dye solutions in use ($\sim 553$ nm and $\sim 567$ nm, respectively). Note that only the line $S_1$ was excited by the dual-broadband-pump scheme in the course of the studies of the CARS noise characteristics.

A high-pressure heatable sample cell (with a heated gas volume of 10 mm in diameter and 100 mm in length) was used. In the experiments at stationary conditions reported here hydrogen was heated up to 1100 K at pressures not exceeding 25 bar. The gas temperature inside the cell was measured by a standard industrial-type thermocouple of Ni-Cr-Ni. The thermocouple, which is protrudes into the gas volume, is chemically stable in a hot hydrogen atmosphere. The thermocouple wires are shielded from direct contact with the hot hydrogen. The hydrogen-filled cell in the reference CARS channel was kept at ambient temperature $T_\mathrm{r} = 296\,\mathrm{K}$. The laser beams were focused into the sample and the reference cells by the same lens, which has a 300-mm focal length. Normally, a USED CARS pump configuration was employed in the experiments. The spatially and spectrally prefiltered CARS beams passed through a monochromator at a small angle to each other and were detected by a diode array, either in a single shot or with accumulation.

## 2 Results and discussion

### 2.1 Temperature measurements and the systematic errors

The version of the 2λ-CARS thermometry realized in our experiments is based on the simultaneous measurement of spectrally integrated energies $I_i$ and $I_j$ of CARS on two separated transitions $i$ and $j$, that are excited over the whole linewidth via lasers with a relatively broad output spectrum. The temperature $T$ is then evaluated from the ratio $I_i(T)/I_j(T)$. This ratio is primarily determined by the population distribution function among the molecular levels, which is Boltzmann at equilibrium conditions. Since two different Stokes lasers are used for simultaneous excitation of the two molecular transitions, a reference channel, with a cell containing the same gas at fixed temperature $T_\mathrm{r}$ and pressure $P_\mathrm{r}$, is installed to normalize the CARS signals. As a result, the normalized ratio $R(T) = [I_i^\mathrm{s}(T)/I_j^\mathrm{s}(T)]/[I_i^\mathrm{r}(T_\mathrm{r})/I_j^\mathrm{r}(T_\mathrm{r})]$ has to be used for the evaluation of the unknown temperature $T$ in the sample channel. The random error of this ratio (and the corresponding error of the temperature) is expected to be reduced even in a single-shot measurement, because the fluctuations in $I^\mathrm{s}$ and $I^\mathrm{r}$ in both the sample and the reference channels of the 2λ-CARS thermometer are supposed to be well correlated when a reasonably adequate beam-interaction geometry is provided in the reference cell with the same resonant gas at stationary conditions.

In the assumption that lasing can be described as a stationary stochastic process with the Gaussian statistics of the fields, the spectrally integrated energy of the CARS pulse can be expressed as follows [12]:

$$I = \mathrm{const} \int \left| X^{(3)}(\omega_1 - \omega_2) \right|^2 I_1(\omega_1) I_2(\omega_2) I_1(\omega_3) \times \delta(\omega_1 + \omega_3 - \omega_2 - \omega_4)\, \mathrm{d}\omega_1\, \mathrm{d}\omega_2\, \mathrm{d}\omega_3\, \mathrm{d}\omega_4\,, \tag{1}$$

where $I_1(\omega)$ and $I_2(\omega)$ are spectral distributions of the energy in the pump and the Stokes laser pulses, $X^{(3)} = X^{(3)}_\mathrm{R} + X^{(3)}_\mathrm{NR}$ is the nonlinear susceptibility,

$$X^{(3)}_\mathrm{R}(\omega_1 - \omega_2) = \sum_k \frac{a_k}{\Omega_k - (\omega_1 - \omega_2) - \mathrm{i}\Gamma_k}\,, \tag{2}$$

$$a_k = \frac{Nc^4}{\mathrm{h}\omega_2^4} \Delta\varrho_k \left[ \frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} \right]_k\,,$$

where $N$ is the number density of the gas molecules, $\Delta\varrho_k$ is the population difference between the initial and the final states of a transition $k$, $\Omega_k$, $(\mathrm{d}s/\mathrm{d}\Omega)_k$ and $\Gamma_k$ are the frequency, the Raman scattering cross section and the linewidth (HWHM) of this transition, respectively. In our experiments the part $I_k$ of the energy of the CARS pulse that is confined within the spectral bandwidth of the resonantly scattered radiation around the transition frequency $\Omega_k$ is measured separately for each of the two probed transitions. In the case of neat hydrogen the contribution from the nonresonant susceptibility $X^{(3)}_\mathrm{NR}$ to this integral CARS energy $I_k$ can be ignored. Because of the large line spacings in the rotational S branch, the contribution of the wings of the neighbouring lines can be neglected as well. Thus, for a given transition k the value of Ik depends on the spectral overlap integral (SOI), which is defined as follows:

$$S(\Gamma_k) = \int \left| \imath\Omega_k - (\omega_1 - \omega_2) - \mathrm{i}\Gamma_k \right|^2 I_1(\omega_1) I_2(\omega_2) I_1(\omega_3) \times \delta(\omega_1 + \omega_3 - \omega_2 - \omega_4)\, \mathrm{d}\omega_1\, \mathrm{d}\omega_2\, \mathrm{d}\omega_3\, \mathrm{d}\omega_4\,. \tag{3}$$

Now the ratio of the CARS pulse energies in the sample and the reference channels can be expressed as:

$$\frac{I_k^\mathrm{s}}{I_k^\mathrm{r}} = \mathrm{const} \frac{N_\mathrm{s}^2}{N_\mathrm{r}^2} \left[ \frac{\Delta\varrho_k^\mathrm{s}}{\Delta\varrho_k^\mathrm{r}} \right]^2 \frac{S(\Gamma_k^\mathrm{s})}{S(\Gamma_k^\mathrm{r})}\,. \tag{4}$$

It should be pointed out that for precise temperature determination, the overlap between the spectra of the lasers and the line of the Raman transition must be so efficient that SOI should represent the integral intensity of the transition line in the CARS spectrum. In the limiting case of a smooth and wide spectral profile of lasers in use (with linewidth $\gg \Gamma$) perfect overlapping takes place and the integral (3) is inversely proportional to $\Gamma$. However, in the real experiment with the stochastic spectral characteristics of lasers the dependence of the SOI on the molecular transition linewidth may differ from this simple relation. To study this problem experimentally the dependence of the SOI on the transition linewidth for $S_1$ and $S_3$ lines has been investigated with various pressures in the sample cell in the range from 1.5 to 25 bar at room temperature and steady-state conditions in the reference cell (see Fig. 1). All the above-mentioned types of dye lasers, with different structures of the radiation spectrum (but always with the spectral width much larger than that of the probed molecular transition) were tried. In all cases a linear dependence of the ratio (4) on pressure was observed. This behavior means that $S \sim 1/\Gamma$, since in the range of hydrogen pressures used in our experiments the spectral lines are homogeneously broadened and $\Gamma = \gamma N$, where $\gamma$ is the collisional broadening coefficient [13]. The results obtained show that the good overlap of the spectra of the lasers and a Raman transition was achieved and experimentally measured CARS energies correctly represent the integral intensities of $S_1$ and $S_3$ lines. Thus, for the normalized ratio of CARS signals for the transitions $S_1$ and $S_3$ one obtains:

$$R \equiv \frac{I_3^s/I_1^s}{I_3^r/I_1^r} = \text{const}\left[\frac{\Delta\varrho_3^s}{\Delta\varrho_1^s}\right]\left[\frac{\Delta\varrho_1^r}{\Delta\varrho_3^r}\right] F_\gamma(T)\,, \qquad (5)$$

where

$$\frac{\Delta\varrho_3^{s,r}}{\Delta\varrho_1^{s,r}} = \frac{X(J=3,\ T^{s,r}) - X(J=5,\ T^{s,r})}{X(J=1,\ T^{s,r}) - X(J=3,\ T^{s,r})}\,,$$

$$X(J,T) = exp[-(BJ(J+1) - DJ^2(J+1)^2)/kT]\,,$$

$$F_\gamma(T) = \frac{\Gamma_1^s(T^s)}{\Gamma_3^s(T^s)}\frac{\Gamma_3^r(T^r)}{\Gamma_1^r(T^r)}\,,$$

$B$ and $D$ are the rotational constants, and $k$ is the Boltzmann constant.

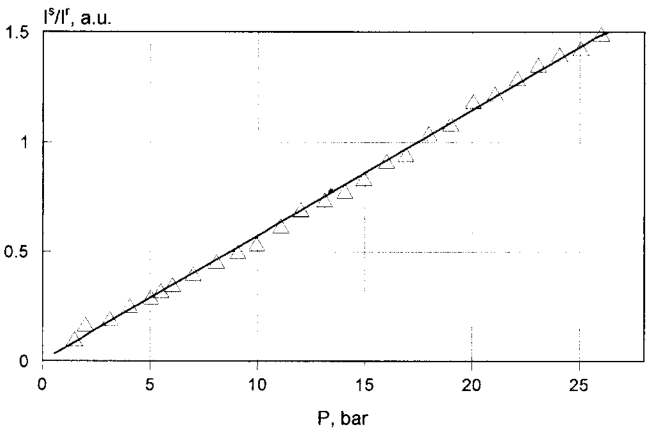


**Fig. 1.** Example of the dependence of the CARS signals ratio $I^s/I^r$ for the $S_1$ transition on the hydrogen pressure in the sample cell: $T_r = 296$ K, the reference cell pressure is 5.5 bar

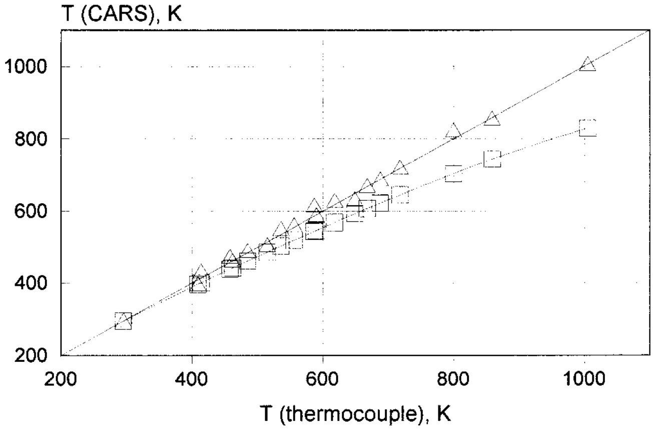


**Fig. 2.** Comparison of temperatures measured by the 2λ-CARS thermometer and the thermocouple: △ – values of the temperature corrected for the $F_\gamma(T)$ temperature dependence. □ – the results of the temperature evaluation if the assumption $F_\gamma(T) = \text{const}$ is made

During the measurements of the temperature the value $R_m$, $m = 1, 2, \ldots$ was determined for each laser shot in a series. Then (5) was solved to provide an estimate $T_m$ of the temperature. The dependence of the $R$ value on the actual gas temperature is determined not only by that of the population differences ratio for a pair of Raman transitions, but also by $J$-dependent variations of the linewidths with temperature. Thus, for correct temperature measurements it is necessary to know how the linewidths or – in case of a linear dependence of linewidths upon the pressure – the collisional broadening coefficients vary with temperature.

Since at present, to our knowledge, there are no published data on the broadening coefficients for the S branch of hydrogen at temperatures higher than 300 K, the measured dependences of the ratios of the integral CARS energies $I_1^s(T)/I_1^r$ and $I_3^s(T)/I_3^r$ on pressure and temperature were used to estimate the corresponding relative variations of the transition linewidths $\Gamma_1(T)$ and $\Gamma_3(T)$. These measurements have been carried out in the range $300\,\text{K} \le T \le 1000\,\text{K}$ at pressures 5.5 and 11 bar in the sample and the reference cells. The gas temperature in the sample cell was measured with the thermocouple. Equation (4) was used afterwards, in the assumption that $S(\Gamma) \sim 1/\Gamma$, to derive $\Gamma^s(T)/\Gamma^r$ for each of the transitions on the basis of calculations of the population differences of the rotational levels and the number densities at a given temperature. Taking into account the temperature dependence of $F_\gamma(T)$ obtained in this way provided satisfactory agreement between the temperature values measured in the independent experiments by the 2λ-CARS thermometer and the thermocouple. In Fig. 2 the corrected values of the temperatures measured by CARS with averaging over 500 laser shots are plotted versus the thermocouple temperatures in the range of 300–1000 K. For comparison, the results of temperature evaluation based on the assumption $F_\gamma(T) = const$ in (5) are also presented in Fig. 2.

## 2.2 *Noise characteristics of single-shot measurements*

Single-shot values of $R_m$ (and hence the estimates $T_m$ of the gas temperature) are random variables. If the evaluated $T_m$ values are assumed to have a Gaussian distribution, the precision of the temperature measurement in each laser shot can be characterized by the relative standard deviation (RSD) $\delta_T$ of the values obtained, which has been estimated on the base of the statistical analysis of the $R_m$ and $T_m$ values in the series of $300-500$ measurements. The value of $\delta_T$ is determined by the RSD of the normalized ratio of CARS signals for a chosen pair of transitions, $\delta_R$, and by the dependence of $R$ on temperature:

$$\delta(T) = \delta_R(T)\left[\frac{T}{R(T)}\frac{\mathrm{d}R(T)}{\mathrm{d}T}\right]^{-1} \equiv \frac{\delta_R(T)}{s(T)}\,, \qquad (6)$$

where $S(T)$ denotes the temperature sensitivity of the 2λ-CARS thermometry. Experimentally the increase in $\delta_T$ with temperature has been observed for different configurations of the laser system (see Fig. 3a,b). It follows from (6) that the increase in $\delta_T$ with temperature can be caused both by the decrease in $s(T)$ and by the increase in $\delta_R$. Each contribution to the $\delta_T$ will be considered separately.

As it is seen from (5) and (6),

$$\frac{T}{R(T)}\frac{\mathrm{d}R(T)}{\mathrm{d}T} = T\left[\frac{\Delta\varrho_\mathrm{s}^2}{\Delta\varrho_\mathrm{r}^2}\right]^{-1}\frac{\mathrm{d}}{\mathrm{d}T}\left[\frac{\Delta\varrho_\mathrm{s}^2}{\Delta\varrho_\mathrm{r}^2}\right] + \frac{T}{F_\gamma(T)}\frac{\mathrm{d}F_\gamma(T)}{\mathrm{d}T} \qquad (7)$$

so that the temperature sensitivity of the 2λ approach is determined, for a given pair of transitions, by the relative derivatives of the ratio of squared population differences (population contribution) and of the function $F_\gamma(T)$ (linewidth contribution). The experimentally obtained values of $s(T) = \delta_R/\delta_T$, together with the calculated temperature dependence of the two contributions to the sensitivity for the transitions $S_1$ and $S_3$, are presented in Fig. 4. The measured values of $F_\gamma(T)$ have been used for the estimate of the linewidth contribution to the $s(T)$. It is clearly seen that the linewidth contribution, as estimated from the experimental data, is much smaller than the population contribution.

With the increase in the temperature, the sensitivity of the 2λ-CARS thermometry, as follows from our estimates, decreases for all the pairs of the S-branch transitions, the strongest decrease being observed for the pair $S_1-S_3$, which was used in our experiments. However, the reason for the choice of this pair is the use of the non-heatable cell in the reference channel, and at room temperature CARS signals at $S_5$ and $S_7$ transitions are very weak and practically not detectable. The installation of a heatable reference cell will allow another pair of transitions to be used, which is assumed to provide a $2-4$ times increase in the precision of temperature measurements.

The experimentally observed increase in $\delta_T$ with temperature exceeds, as shown in Fig. 3, that expected from the decrease in the sensitivity. The additional contribution to $\delta_T$ is supposed to be caused by the increase in the factor $\delta_R(T)$ in (6). The value of $\delta_R$ is determined mainly by shot-to-shot fluctuations of the two pairs of statistically correlated random values of CARS energies $I^\mathrm{s}$ and $I^\mathrm{r}$, depending on the spectral profiles of the pump lasers, the laser intensities and their distribution across the beam, and a spatial overlap of the interacting beams. If we assume that the contributions to the CARS energies determined by spatial parameters (SPAT) and spectral parameters (SPEC) are multiplicative ($I = I_\mathrm{SPAT}I_\mathrm{SPEC}$) and statistically independent, the RSD of the measured CARS signals can be written as follows:

$$\delta^2_\mathrm{CARS} = \delta^2_\mathrm{SPAT} + \delta^2_\mathrm{SPEC}\,, \qquad (8)$$

where $\delta_\mathrm{SPAT}$ and $\delta_\mathrm{SPEC}$ are determined by fluctuations of spatial characteristics of the CARS process and spectral pa-

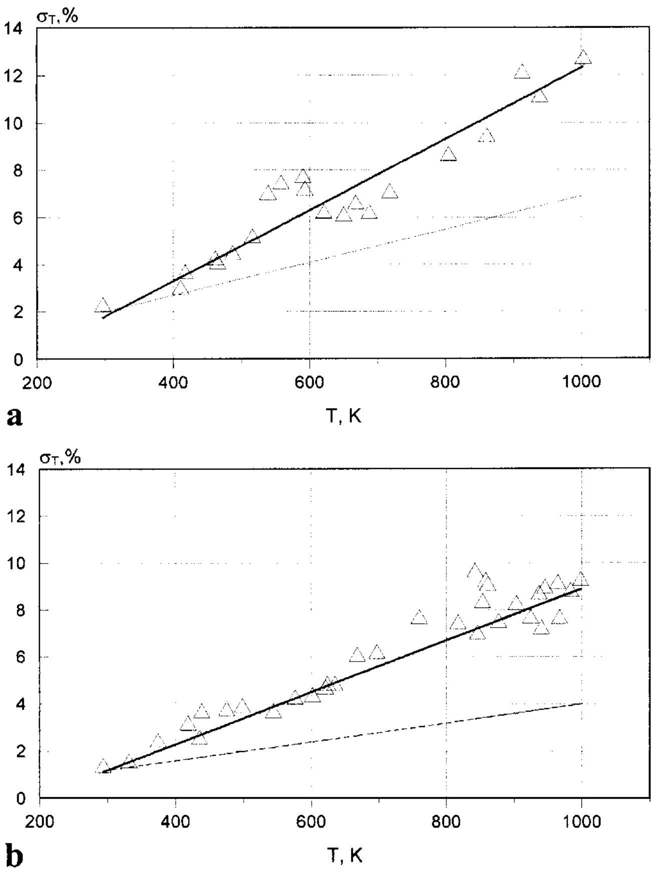


**Fig. 3a,b.** Experimentally estimated relative standard deviation $\delta_T$ of the temperature values measured with the DFDL (**a**) and the SLDL (**b**) configurations. Dashed lines represent the calculated behavior of $\delta_T$ due to variation of the sensitivity factor alone, solid lines are guides for the eye

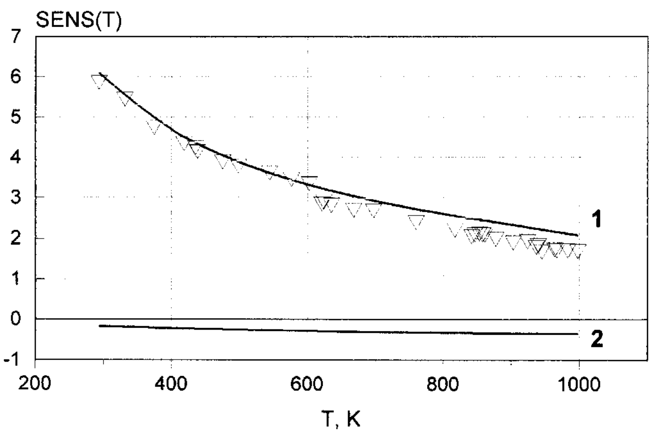


**Fig. 4.** Experimentally measured values of the sensitivity (▽ ) and estimated different contributions to the sensitivity for the $S_1-S_3$ pair of transitions (*solid lines*). 1 – relative derivative of the ratio of the squared population differences; 2 – relative derivative of the function $F_\gamma(T)$

rameters (SOI), respectively. The small noise of the detector sensitivity (RSD $\approx 3\%$) is neglected in this consideration.

The ratio of the CARS signals in the sample and the reference channels for a given transition also has a random noise contribution. The RSD $\delta_{sr}$ of the sample-to-reference ratio (4), if similar laser-beam interaction conditions in the sample and the reference cells are assumed, is mainly determined by the correlation of the random values of SPEC:

$$\delta_{sr}^2 = (\delta_{SPEC}^s)^2 + (\delta_{SPEC}^r)^2 - 2\delta_{SPEC}^s \delta_{SPEC}^r \varrho \tag{9}$$

where $\delta_{SPEC}^s$ and $\delta_{SPEC}^r$ denote the RSD for $I^s$ and $I^r$, accordingly, and $\varrho$ is the correlation coefficient ($|\varrho| \leq 1$). The value of $\delta_{sr}^2$ can vary from $(\delta_{SPEC}^s)^2 + (\delta_{SPEC}^r)^2$ (total independence, $\varrho = 0$) down to $(\delta_{SPEC}^s - \delta_{SPEC}^r)^2$ (perfect correlation, $\varrho = 1$).

In order to understand the behaviour of $\delta_R$ with temperature the dependences of the contribution $\delta_{SPEC}$ to the RSD of the CARS signals $\delta_{CARS}$ and of the RSD of the sample-to-reference ratio $\delta_{sr}$ on the molecular transition linewidth were studied for both $S_1$ and $S_3$ lines by using pump lasers with different spectral characteristics. The transition linewidths varied by increasing the hydrogen pressure in the sample cell from 1.5 to 25 bar at room temperature, the widths being linearly related to pressure in this range [13]. As an example, Fig. 5 shows the dependence of $\delta_{SPEC}$ on the linewidth of the $S_1$ transition for three configurations of the dye laser module: DFDL, dual broadband (two broadband dye lasers), and AS-EDL. For all the configurations the common tendency to a decrease in $\delta_{SPEC}$ with an increase in the transition linewidth is observed, which can be explained by the increase in the number of spectral components of the laser radiation being averaged in the CARS process. The DFDL configuration is characterized by larger values of $\delta_{SPEC}$, and the dual broadband is characterized by smaller ones. In these two cases the approximation by a power law shows similar $\delta_{SPEC}$ dependence on the transition linewidth ($\delta_{SPEC} \sim 1/\Gamma$), which is supposed to be explained by the same mode structure of dye laser radiation (with a mode spacing of about $0.03\,\mathrm{cm}^{-1}$). The decrease in the absolute value of $\delta_{SPEC}$ in the case of a dual broadband configuration should then be ascribed to a larger number of dye laser modes contributing to CARS signal generation. In the case of the SLDL configuration, a decrease in $\sigma_{SPEC}$ has a weaker dependence on the linewidth ($\delta_{SPEC} \sim 1/\Gamma^{0.7}$).

The variation in $\sigma_{sr}$ with the ratio $\widetilde{\Gamma} = \Gamma^s/\Gamma^r$ for the linewidths of the $S_1$ transition in the sample and the reference cells, measured in the pressure range of 1.5–25 bar, is shown in Fig. 6. All three curves, obtained for different configurations of the dye laser module, have similar general features: a minimum, corresponding to equal transition linewidths in both cells; a steep growth, when the linewidth in the sample cell is decreasing relative to that in the reference cell, and a slow growth in the opposite case. The observed behavior of $\delta_{sr}$ could be reasonably well described on the basis of (9). If the difference between the transition linewidths in the sample and the reference cells is large, the numbers of spectral components of the laser radiation contributing to the formation of the CARS signals are also different, so that uncorrelated components of anti-Stokes radiation appear in the channel with the larger transition linewidth. This results in the reduction of the correlation coefficient $\varrho$ and the corresponding growth of $\delta_{sr}$. At the same time, the value of $\delta_{SPEC}^s$ increases with the decrease of the transition linewidth (see Fig. 5). Thus, when $\widetilde{\Gamma} < 1$, both factors lead to an increase in $\delta_{sr}$ resulting in its strong dependence on $\widetilde{\Gamma}$, whereas when $\widetilde{\Gamma} > 1$ the two factors contribute in the opposite directions and the dependence of $\delta_{sr}$ on $\widetilde{\Gamma}$ becomes weaker.

For all the dye laser configurations the dependences of $\delta_{sr}$on the ratio of the transition linewidths $\widetilde{\Gamma}$ are found to be similar to each other if the values $\delta_{sr}$ in each of the three data sets are reduced by the corresponding minimal value. The minimal values of $\delta_{sr}$ characterize the best correlation of the random values $I_s$ and $I_r$ and are mainly determined not by the dye laser configuration, but by the provided similarity of the laser-beam interaction conditions in the sample and the reference channels, as well as by the noise of the detector. The lower limit of $\delta_{sr}$ for our set-up was about $3-4\%$ as estimated by measuring $\delta_{sr}$ for two parts of the same pump beam after it passed simultaneously through the sample and through the reference channels. The minimal value of $\delta_{sr}$ is rather critical to the adjustment of the CARS system and may vary from experiment to experiment. So it is worth mentioning that

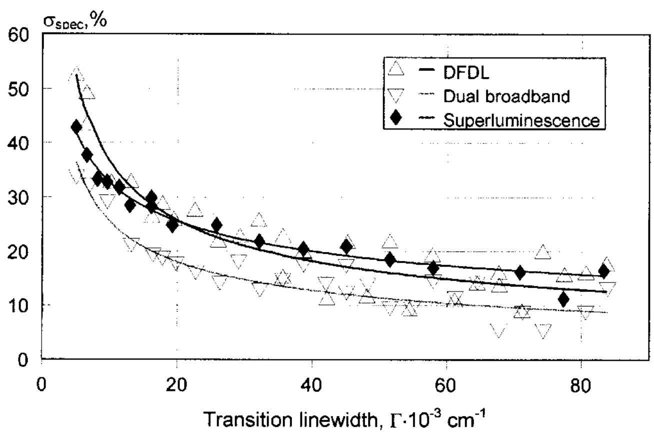


**Fig. 5.** The dependence of a spectral contribution to the RSD of CARS signal on the linewidth of the $S_1$ transition for different configurations of the dye laser module. Symbols mark the experimental data, solid lines show the approximation by a power law

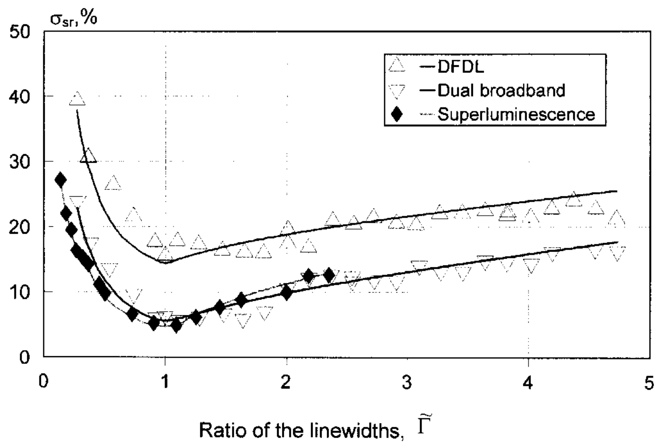


**Fig. 6.** The dependence of RSD for the sample-to-reference ratio from the ratio of the linewidths of the $S_1$ transitions in the sample and the reference cells for different configurations of the dye laser module. Symbols mark the experimental data, solid lines show their approximation by smooth curves

in experiments not presented in Fig. 6 the minimal value of $\delta_{sr} \approx 5\%$ was achievable also for the DFDL configuration.

Our experiments showed the $1.5-2.5$ times enhancement of $\delta_R$, as well as of $\delta_{sr}$, for each of the transitions, during heating of the gas. The evaluation of the temperature dependence of the broadening coefficients for the $S_1$ and $S_3$ lines, based on the published data [14, 15] and our measurements, leads to the conclusion that the linewidths of both transitions decrease at isobaric heating. This brings, in accordance with Fig. 6, us to the observed increase in $\delta_{sr}$ for each of the transitions and, as a result, to the corresponding increase in $\delta_R$.

## 3 Conclusions

The analysis of the precision is presented of a single-shot 2λ-CARS thermometry employing the pure rotational transitions $S_1$ and $S_3$ of hydrogen. The temperatures measured with averaging over 500 laser shots ($300\,K < T < 1100\,K$, pressures of 5.5 and 11 bar) agree with the thermocouple measurements within the accuracy of $3-5\%$. The RSD of single-shot temperature measurements $\delta_T$ varied from $\sim 1.5\%-2\%$ at room temperature to $\sim 8\%$ (ASEDL) and 12% (DFDL) at 1100 K. The observed growth of $\delta_T$ with an increase in the temperature is explained by the simultaneous influence from two factors: a decrease in the sensitivity to temperature variations of the intensity ratio for the pair of $S_1-S_3$ lines, and a enhancement of the RSD of the normalized ratio $\delta_R$ due to decrease in the correlation of CARS signals in the sample and the reference channels for both transitions.

The comparative analysis of noise characteristics of the CARS signals, obtained for different configurations of the dye laser module, showed that the lowest noise level was achieved with the ASEDL configuration. The level of this noise was similar to that achieved with the dual broadband CARS setup. For all the configurations in use $\delta_{SPEC}$ was inversely proportional to the transition linewidth, and a decrease of the noise at higher pressures was observed. It should be noted that the RSD $\delta_{sr}$ of the sample-to-reference ratio is not so critical to the spectral characteristics of the dye laser module.

The reduction of $\delta_{sr}$ was found when the transition linewidths in the sample and the reference channels were equal. It means that in the course of 2λ-CARS thermometry in a real combustor the pressure in the reference channel should be chosen so that in the sample channel the conditions of broadening of the transitions employed were in average identical.

On the base of the data analyzed in the present paper, the ASEDL configuration has been chosen for future 2λ-CARS single-shot temperature measurements in the technical combustor. In this case the precision of about 5–6% at temperatures 1000–2000 K is expected to be reached.

## References


1. D.A. Greenhalgh: In *Advances in Nonlinear Spectroscopy*, Vol.15, ed. by R.J.H. Clark, R.E. Hester (John Wiley & Sons, Chichester 1988) p.193-251
2. M. Alden, P.-E. Bengtsson, H. Edner, S. Kröll, D. Nilsson: Appl.Opt. **28**, 1068 (1989)
3. W. Stricker, M. Woyde, R. Lückerath, V.Bergmann: Ber. Bunsenges. Phys. Chem. **97**, 1608 (1993)
4. P. Snowdon, S.M. Skippon, P. Ewart: Appl. Opt. **30**, 1008 (1991)
5. W. Clauss and R. Söntgen: Single shot CARS in H2. Diagnostics Group Meeting DLR, Stuttgart, April 16, 1991
6. M.A. Alden, K. Fredriksson, S. Walli: Appl. Opt. **23**, 2053 (1984)
7. M. Péalat, M. Lefebvre: Appl. Phys. B **53**, 23 (1991)
8. W. Clauss, A.A. Ilyukhin, D.N. Kozlov, V.V. Smirnov, O.M. Stelmakh, K.A. Vereschagin: Appl. Phys. B **62**, 279 (1996)
9. R. Beach, S.R. Hartman: Phys. Rev. Lett. **53**, 663 (1984)
10. P. Ewart: Opt. Commun. **55**, 124 (1985)
11. M. Alden, P.-E. Bengtsson, H. Edner, S. Kröll, D.Nilsson: Appl. Opt. **28**, 3206 (1989)
12. M.A. Yuratich: Mol. Phys. **38**, 625 (1979)
13. R.A.J. Keijser, J.R. Lombardi, K.D. Van Den Hout, B.C. Sanctuary, H.F.P. Knaap: Physica **76**, 85 (1974)
14. L.A. Rahn, R.L. Farrow: Phys. Rev. A **43**, 6075 (1991)
15. L.A. Rahn, G.J. Rosasco: Phys.Rev. A **41**, 3698 (1990)