Measurements of the local temperature correlation time in a turbulent flame using coherent anti-Stokes Raman spectroscopy

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Abstract. The feasibility to determine the timescale of pulsations of "instantaneous" local temperatures in a turbulent flame at a microsecond time scale by using coherent anti-Stokes Raman scattering (CARS) spectroscopy is demonstrated for the first time to our knowledge. A laboratory laser measurement complex was utilized, based on two CARS-spectrometers employing synchronized pulse-repetitive lasers with 10 ns pulse duration. The system enabled to record, with high temporal resolution (in one single laser shot) and at a variable delay between two sequential shots following each other in pairs at a repetition rate of 10 Hz, series of CARS spectra of N\textsubscript{2} molecules from a probe volume as small as 0.03×0.03×2 mm\textsuperscript{3}. From the spectra, "instantaneous" temperatures at a given delay were derived. The obtained values enabled calculation of the correlation coefficient of temperature pulsations vs the delay. The results are presented for the series of 500 single-shot coupled measurements, at the delays in the range 1 μs – 0 ms, of local gas temperatures in a few points of an open turbulent partially premixed methane-air flame of a model burner with visually distinguishable stability of combustion. The average temperatures were between 1500 K and 1800 K. The measurements allowed temperature correlation times in the selected points of the flame to be derived.

1. Introduction
Development of non-contact methods and equipment for high spatial and temporal resolution optical diagnostics of turbulent combustion, which provide previously unavailable characteristics of physico-chemical and gas-dynamic processes, is an urgent task, directly related to design and development of combustion chambers for advanced gas turbine engines. Coherent anti-Stokes Raman scattering (CARS) spectroscopy [1–4] is a developed technique to determine local gas temperatures in reactive flows. CARS is a process of parametric four-wave mixing in a medium, when coherent pump and lower-frequency (Stokes) electromagnetic waves generate radiation at a higher (anti-Stokes) frequency. The efficiency of the mixing increases by several orders of magnitude under the conditions of resonant interaction of the radiation with probe molecules at frequencies of their Raman-active transitions. It is these transitions which are employed in diagnostic applications of CARS. Focusing of the pump and Stokes laser beams into a small probe volume ensures high spatial resolution of the measurements.
Local temperatures of nitrogen-containing gas media can be derived from CARS spectra of N\textsubscript{2} molecules, each spectrum registered during a single shot of a high-energy pulsed laser ($\approx 10$ ns). The single-shot measurement can be regarded as "instantaneous" in relation to characteristic times of thermodynamic parameter variations of the probed medium. Crossing the beams which are provided by two similar CARS-spectrometers, with temporally synchronized repetitive laser pulses of nanosecond duration, in the focus and recording the spectra by each of the spectrometers in a single laser shot at different delays between these shots may allow the timescale of temperature pulsations to be determined.

The aim of the present study is to demonstrate the feasibility to derive the time of correlation of "instantaneous" local temperatures in a turbulent flame at a microsecond scale by using CARS-spectroscopy.

2. Experiment

For measurements of local temperatures in a flame CARS-spectra of rovibrational transitions of N\textsubscript{2} molecules, with well-known spectroscopic parameters, were employed. In the four-wave mixing process of CARS in a medium with optical nonlinearity the waves of the pump and Stokes radiations, at frequencies $\nu_p$ and $\nu_s$, generate a wave of radiation at the higher frequency, $\nu_{as} = 2\nu_p - \nu_s$, shifted to the anti-Stokes range. Under resonance conditions, the frequency difference ($\nu_p - \nu_s$) is close to the frequency $\nu_R$ of a Raman-active transition of the probed molecules: $\nu_p - \nu_s \approx \nu_R$ (see a schematic in figure 1).

![Figure 1. Schematic of CARS process involving transitions between rotational levels of the ground ($\nu = 0$) and the first excited ($\nu' = 1$) vibrational states of N\textsubscript{2} molecules at broadband Stokes pumping; $J$ and $J'$ are rotational quantum numbers.](image)

The experimental setup consisted of a laser measurement complex based on two synchronized laboratory CARS-spectrometers and a model burner (see figure 2).

The two spectrometers allowed independent two-channel single-shot registration of CARS-spectra from the same spatial point to be performed, meanwhile the delay between the shots (the moments of the measurement) could be varied within a broad range at a microsecond scale. Each of the spectrometers (see the detailed description in [5]) contained a laser module, with a high-energy narrowband frequency-doubled pulse-repetitive Q-switched Nd\textsuperscript{3+}:YAG laser ($\lambda_p = 532$ nm, 130 mJ, 10 ns, 10 Hz) used as the pump laser. Part of its radiation ($\approx 100$ mJ) was split to pump a dye laser employed as a source of broadband Stokes radiation ($\lambda_s \approx 607$ nm, $\Delta \lambda_s \approx 6$ nm). Nanosecond pump and Stokes laser pulses were temporally overlapped and were focused into the same probe volume by a $f = 300$ mm focal length lens L1 in a 3-D BOXCARS phase-matching configuration [6]. This configuration determined small dimensions of the probe volume ($\approx 0.03 \times 0.03 \times 2$ mm$^3$), defined by the beam overlap in the focal region, i.e. the high spatial resolution of the measurements.

Non-linear four-wave mixing of the laser radiations amplified by Raman-active rovibrational resonances of N\textsubscript{2} molecules with Raman shifts $\approx 2240$-2330 cm$^{-1}$ in the $Q$-branches of the spectrum...
\((v \rightarrow v' = v + 1, \ v = 0, \ 1, \ J \rightarrow J' = J)\) resulted in generation in the probe volume of two spatially separated beams of the detected anti-Stokes radiation at wavelengths near \(\lambda_{as} = \lambda_p/(2 - \lambda_p/\lambda_s) \approx 473 \text{ nm}\). Each beam of the anti-Stokes radiation was collimated by a lens L2, spectrally decoupled from the scattered light of the pump and Stokes lasers by a bandpass interference filter, and directed to the entrance slit of a spectrograph (1000-mm focal length, 2400 lines/mm grating) of the corresponding CARS-spectrometer. Spectra were being recorded by an intensified gated 2-D CCD detector, triggered synchronously with the laser shot of the spectrometer.

Synchronization in time of repetitive nanosecond laser shots of the two CARS-spectrometers and adjustment of the delay between the pairs of these shots in the range of 0.1 \(\mu\text{s} - 10 \text{ ms}\) was arranged via external triggering of the pump lasers by using a delay and pulse generator.

**Figure 2.** Scheme of the experimental setup; L1, L2 – focusing and collimating lenses.

A model burner, with a system of mass flow controllers for fuel and oxidizer, represented an open continuously operating partially premixed methane-air torch. A combustible mixture with a total volume flow rate of \(\approx 0.45 \cdot 10^{-3} \text{ m}^3/\text{s}\) was supplied through a central duct of the burner (5.5 mm in diameter) and a coaxial slit (1-mm width, 18 mm in diameter). The mixture flow velocity at the central duct exit was about 10 m/s, and the height of the main flame reached \(\approx 70 \text{ mm}\). The slit was providing a stabilizing circular auxiliary flame around the main flame kernel, with the mixture flow velocity lower than in the central duct. The design of the burner ensured the axial symmetry of the flame. The burner was installed onto a moveable mount which enabled flame positioning as related to the diagnostics probe volume with an accuracy of \(\approx 1 \text{ mm}\).

The choice of the flame regions with relatively stable or stochastically varying temperatures, in which the probe volume could be placed during the measurements, was based on "instantaneous" images of the flame chemiluminescence registered during a short exposure time (50 \(\mu\text{s}\)) with a period of 100 ms by using a gated CCD camera. The measurement points were selected at the flame axis.

The analysis of the recorded spectral profiles to derive the temperatures was being carried out by using a special fitting code, implementing the approximation of each of them by selecting a theoretical profile, which minimized the deviations of the two profiles, from the library of the spectra [8]. The library spectra were to be pre-calculated by the code on the basis of known molecular constants for a few sets of variable parameters of the theoretical model. The \(\text{N}_2\) CARS-spectra registered by each of the spectrometers were distorted, as compared to the theoretical ones, by the Stokes laser characteristic broad spectral profile which randomly fluctuates from one laser shot to another. To eliminate the systematic part of the distortion, the Raman-resonant spectra obtained in each of the registration
channels, were normalized by the average spectral profile of the corresponding Stokes laser represented by an accumulated in 100 laser shots non-resonant spectrum of coherent anti-Stokes scattering in a cell filled with Ar. The "instantaneous" gas temperature in the probe volume was derived as a value of the respective variable parameter of the theoretical spectrum which was best-fitting the normalized experimental spectrum.

3. Results
In the course of the measurements, the recordings of two series of 500 single-shot N₂ CARS-spectra following in 100 ms were being accomplished in parallel using both CARS spectrometers of the measuring complex. In doing so, for each pair of the series a definite delay \( \tau \) between the pulses of the two pump lasers was set in the range from 1 \( \mu \)s to 10 ms.

In the spatial point of the flame with the relatively stable temperature, located in the zone of combustion products, only one pair of series was recorded, at zero delay between the pump laser pulses. In each of the series, the spectra were characterized by high reproducibility in the profile and amplitude, that refers to stability of the gas medium within the probe volume during the time of a series registration (\( \approx 1 \) min). In the point of the flame with the pulsating temperature, located closer to the edge of the burner, in the vicinity of the flame front, the pairs of the series of spectra with various delays between the laser shots were recorded.

Using sets of \( N \) values of "instantaneous" local temperatures \( T_{i,2}, i = 1,\ldots, N \) is a serial number of the value, obtained in each channel of registration, histograms of the frequency distributions of temperatures were plotted, the average values \( T_{av,1,2} \) and the standard deviations \( \sigma_{av,1,2} \) of the single-shot temperatures were evaluated, and the sample Pearson correlation coefficients \( r_T \) of the temperatures derived in the two channels were calculated, in dependence on the delay \( \tau \) between the pump laser pulses, according to the known relation:

\[
r_T(\tau) = \frac{\langle \Delta T_{i,1}(0) \cdot \Delta T_{i,2}(\tau) \rangle}{\sqrt{\langle \Delta T_{i,1}^2 \rangle \cdot \langle \Delta T_{i,2}^2 \rangle}},
\]

where \( \Delta T_{i,1,2} \) are the differences \( (T_{i,1,2} - T_{av,1,2}) \) between the "instantaneous" and the average temperatures for the respective registration channel, and \( \langle \cdots \rangle \) means averaging over all \( N \) values in a set.

In the pair of temperature sets obtained in the point with the stable temperature, at \( \tau = 0 \) \( \mu \)s, the average values were calculated to be \( T_{av,1} \approx 1676 \) K and \( T_{av,2} \approx 1692 \) K with standard deviations (SD) \( \sigma_{av,1} = 55 \) K (\( \delta T_1 = 3.3\% \)) and \( \sigma_{av,2} = 72 \) K (\( \delta T_2 = 4.3\% \)), respectively. Frequency distributions of these temperatures in the sets (see figure 3) are close to the normal ones as shown by the dotted line. The SD

![Figure 3. Frequency distributions of local gas temperatures derived from two series of single-shot CARS spectra in the flame point with relatively stable temperature at delay \( \tau = 0 \) \( \mu \)s; the dotted line shows the approximation by the normal distribution: (a) in channel 1, (b) in channel 2.](image-url)
values in this case characterize primarily random and non-correlated deviations of temperature values, derived in each of the registration channels, due to both shot-to-shot fluctuations of broad spectral profiles of the Stokes dye lasers and errors of temperature evaluation from the spectra introduced by the numerical approximation procedure. As a consequence, the value of the correlation coefficient was found to be only \( r_T(0) = -0.07 \). Thus, the derived SD values characterize the instrumental measurement errors, specific to each of the spectrometers, and are comparable to that found previously in [7] for the similar spectrometer. These results demonstrate the stability of the operation both of each of the spectrometers and of the measurement complex as a whole.

\[
\sigma_{av} = 350 \text{ K} \quad (\delta T_1 = 23\%) \quad \text{and} \quad \sigma_{av} = 440 \text{ K} \quad (\delta T_2 = 30\%),
\]

respectively, at the discrepancy of the average values in 36 K, or 2%, and become significantly larger than the corresponding instrumental error of single-shot measurements. In this point, the frequency distributions of the temperatures differ from the normal ones in both channels (see figure 3) and show a significant amount of the reduced temperature values in the flow. In this case the SD values characterize average amplitudes of temperature pulsation amplitudes due to flow turbulence.

The examples of frequency distributions of single-shot local gas temperatures in the flame point with pulsating temperature are presented in figure 4. In this point, the average values in the pair of sets are slightly lower and equal to \( T_{av} = 1515 \text{ K} \) and \( T_{av} = 1479 \text{ K} \). However, the SD values increase by almost an order of magnitude to reach \( \sigma_{av} = 350 \text{ K} \) \( (\delta T_1 = 23\%) \) and \( \sigma_{av} = 440 \text{ K} \) \( (\delta T_2 = 30\%) \), respectively, at the discrepancy of the average values in 36 K, or 2%, and become significantly larger than the corresponding instrumental error of single-shot measurements. In this point, the frequency distributions of the temperatures differ from the normal ones in both channels (see figure 3) and show a significant amount of the reduced temperature values in the flow. In this case the SD values characterize average amplitudes of temperature pulsation amplitudes due to flow turbulence.

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The value of the correlation coefficient of the temperatures in the two channels was found to be equal to \( r_T = 0.88 \) at \( \tau = 0 \) \( \mu \text{s} \). With the increase of the delay in the range of 1 \( \mu \text{s} \) – 10 ms its value starts to decrease at \( \tau \geq 10 \) \( \mu \text{s} \) (see figure 5). The correlation time derived as the time constant of the exponential decay curve fitted to the values obtained from the experiment: \( r_T(\tau) = r_0 \cdot \exp (-\tau/\tau_0) \), shown in figure 4 by a solid line and characterizing the timescale of temperature pulsations, appeared to be equal in this point of the flame to \( \tau_0 = 126 \pm 18 \) \( \mu \text{s} \).

4. Conclusion

Employing the laboratory laser measurement complex, based on two CARS-spectrometers with synchronized high-energy pulse-repetitive lasers with 10 ns pulse duration, and varying a temporal delay between two sequential laser shots in the microsecond range, the feasibility of measurements of the timescale and of the average amplitude of instantaneous local gas temperature pulsations in a turbulent flame was demonstrated, for the first time to our knowledge. In the partially premixed flame of the model open methane-air burner with continuous operation, temperatures and time of their correlation were derived in spatial points which differ in stability of combustion.

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