Time-resolved measurement of the local equivalence ratio in a gaseous propane injection process using laser-induced gratings

Thomas Seeger, Johannes Kiefer, Markus C. Weikl and Alfred Leipertz

Lehrstuhl für Technische Thermodynamik, Friedrich-Alexander Universität Erlangen-Nürnberg,
Am Weichselgarten 8, 91058 Erlangen, Germany
ts@ltt.uni-erlangen.de

Dimitrii N. Kozlov

A.M. Prokhorov General Physics Institute, Russian Academy of Sciences,
Vavilov str. 38, 119991 Moscow, Russia
dnk@kapella.gpi.ru

Abstract: For the first time laser-induced gratings (LIGs) have been used for the investigation of a non-stationary pulse-repetitive injection process of gaseous propane, C_3H_8, into air. By recording and evaluating single-shot LIG signals it was possible to determine, on a cycle-averaged basis, the temporal evolution of the local (within a probe volume 300 µm in diameter and 10 mm in length) equivalence ratio and by this the fuel-air ratio. Two different data treatment strategies, subject to C_3H_8 concentration range, were first tested at stationary conditions and then used to evaluate the LIG signals obtained during the injection process. The relative standard deviation of single-shot measurements were estimated to be 0.14 and 0.32 at 0.8 % and 10 % of propane concentration, respectively.

OCIS codes: (190.1900) Diagnostic applications of nonlinear optics; (120.1740) Combustion Diagnostics.

References and links

1. H. J. Eichler, P. Günter, and D. W. Pohl, "Laser-Induced Dynamic Gratings," Springer Verlag, Berlin (1986).
2. A. Stampanoni-Panariello, D. N. Kozlov, P. P. Radi, and B. Hemmerling, "Gas phase diagnostics by laser-induced gratings I. Theory," Appl. Phys. B. **81**, 101-111 (2005).
3. A. Stampanoni-Panariello, D. N. Kozlov, P. P. Radi, and B. Hemmerling, "Gas phase diagnostics by laser-induced gratings II. Experiments," Appl. Phys. B. **81**, 113-129 (2005).
4. A. Stampanoni-Panariello, B. Hemmerling, and W. Hubschmid, "Temperature measurements in gases using laser-induced electrostrictive gratings," Appl. Phys. B **67**, 125-130 (1998).
5. R. C. Hart, R. J. Balla, and G. C. Herring "Nonresonant referenced laser-induced thermal acoustics thermometry in air," Appl. Opt. **38,** 577-584 (1999).
6. E. B. Cummings, "Laser-induced thermal acoustics: simple accurate gas measurements," Opt. Lett. **19,** 1361-1363 (1994).
7. A. Stampanoni-Panariello, B. Hemmerling, and W. Hubschmid, "Electrostrictive generation of nonresonant gratings in the gas phase by multimode lasers," Phys. Rev. A **51**, 125-130 (1998).
8. B. Hemmerling, W. Hubschmid, and A. Stampanoni-Panariello, "Temperature and mixture fraction measurements in gases by laser-induced electrostrictive grating," Proc. Combust. Inst. **27,** 69-75 (1998).
9. B. Hemmerling and D. N. Kozlov, "Generation and temporally resolved detection of laser-induced gratings by a single, pulsed Nd:YAG laser," Appl. Opt. **38**, 1001-1007 (1999).
10. B. Hemmerling, M. Neracher, D. Kozlov, W. Kwan, R. Stark, D. Klimenko, W. Clauss, and M. Oschwald, "Rocket nozzle cold-gas flow velocity measurements using laser-induced gratings," J. Raman Spectrosc. **33,** 912-918 (2002).
11. D. J. W. Walker, R. B. Williams, and P. Ewart, "Thermal grating velocimetry," Opt. Lett. **23,** 1316-1318 (1998).

12. R. Stevens and P. Ewart, "Single-shot measurement of temperature and pressure using laser-induced thermal gratings with a long probe pulse," Appl. Phys. B **78,** 111-117 (2004).
13. R. Stevens and P. Ewart, "Simultaneous single-shot measurement of temperature and pressure along a one-dimensional line by use of laser-induced thermal grating spectroscopy," Opt. Lett. **31,** 1055-1057 (2006).
14. B. Hemmerling, D. N. Kozlov, and A. Stampanoni-Panariello, "Temperature and flow-velocity measurements by use of laser-induced electrostrictive gratings," Opt. Lett. **25,** 1340-1342 (2000).
15. D. N. Kozlov, "Simultaneous characterization of flow velocity and temperature fields in a gas jet by use of electrostrictive laser-induced gratings," Appl. Phys. B **80,** 377-387 (2005).
16. M. S. Brown and W. L. Roberts, "Single-point thermometry in high-pressure, sooting, premixed combustion environment," J. Propul. Power **15**, 119-130 (1999).
17. H. Latzel, A. Dreizler, T. Dreier, J. Heinze, M. Dillmann, W. Stricker, G. M. Loyd, and P. Ewart, "Thermal grating and broadband degenerated four-wave mixing spectroscopy of OH in high-pressure flames," Appl. Phys. B. **67**, 667-673 (1998).
18. M. S. Brown, Y. Li, W. L. Roberts, and J. R. Gord, "Analysis of transient-grating signals for reacting-flow applications," Appl. Opt. **42**, 566-578 (2003).
19. B. Hemmerling, D. N. Kozlov, O. M. Stel'makh, and B. Attal-Trétout, "Diagnostics of water-containing gas mixtures using thermal laser-induced gratings," Chem. Phys. **320**, 103-117 (2006).
20. M. C. Weikl, F. Beyrau, J. Kiefer, T. Seeger, and A. Leipertz, "Combined coherent anti-Stokes Raman spectroscopy and linear Raman spectroscopy for simultaneous temperature and multiple species measurements," Opt. Lett. **31**, 1908-1910 (2006).
21. T. Seeger, J. Jonuscheit, M. Schenk, and A. Leipertz, "Simultaneous Temperature and Relative Oxygen and Methane Concentration Measurements in a Premixed Sooting Flame Using a Novel CARS-Technique," J. Mol. Struct. **661-662**, 515-524 (2003).
22. F. Beyrau, M. C. Weikl, I. Schmitz, T. Seeger, and A. Leipertz, "Locally resolved investigation of the vaporization of GDI sprays applying different laser techniques," Atom. Sprays **16**, 319-330 (2006).
23. B. Hemmerling and D.N. Kozlov, "Collisional relaxation of singlet O_2 (b $^1\Sigma^+_g$) in neat gas investigated by laser-induced grating technique," Chem. Phys. **291**, 213-242 (2003).

1. Introduction

Diffraction by laser-induced gratings (LIG) is a well known technique for the investigation of thermophysical properties of liquids and condensed phase matter [1]. In recent years a lot of effort has been put to demonstrate the potential of LIGs for gas-phase diagnostics. In general, laser-induced gratings are the spatially-periodic modulations of the refractive index, which can be induced by two interfering pump laser beams. In the gas-phase there are two dominating contributions to such gratings, which are fundamentally different. These contributions can be generated by electrostriction or by collisional relaxation of the absorbed pump laser radiation, and are called electrostictive or thermal gratings, respectively. The overall modulation of the refractive index results from their interference. The measured signal is the part of a third, directed at an appropriate angle, laser beam Bragg-diffracted by this modulation. Electrostrictive gratings are generated at any frequency of the pump laser light. In media that weakly absorb pump radiation electrostriction provides the major contribution to the signal. In stronger absorbing media diffraction by thermal gratings may appear as the dominant process. More information about the theory of LIGs and their application to gas-phase diagnostics can be found, e. g., in recent reviews [2, 3].

In non-reacting flows the LIGs technique has already been used to accomplish local temporally-resolved measurements of the gas temperature (e. g. [4, 5]), gas dynamic properties (e. g., [6, 7]), species concentration in binary isothermal gas mixtures (e. g., [8, 9]) or flow velocity (e. g., [10, 11]). Furthermore simultaneous measurements of temperature and pressure [12, 13] or temperature and flow velocity [14, 15] have been performed successfully. This technique has also been applied to flames for temperature measurements (e. g., [16-19]).

All the results obtained show that the LIGs technique is a promising method for the investigation of mixture formation in injection and combustion processes. Up to now for the investigation of these processes both linear and non-linear Raman techniques have often been used (e. g., [20, 21]). However, because of the small Raman scattering cross sections the signal levels are comparatively low, which limits the sensitivity of the methods in this application [20, 21]. In the present work the LIGs technique has been applied to study

quantitatively the injection of gaseous propane, C_3H_8, into air under non-reacting conditions. The technique was used for the first time, to the best of our knowledge, to determine the temporal evolution of the local equivalence ratio in a non-stationary pulse-repetitive flow on a cycle-averaged basis. Propane is one of the main components of liquefied petroleum gas (LPG), which is of special interest as an alternative fuel in spark-ignited engines. In general, the LIGs technique is also applicable to other gaseous fuels prior to ignition. For concentration measurements only a proper pump laser wavelength is required in order to achieve resonant excitation of species to be detected.

2. Experimental setup

A scheme of the experimental setup is shown in Fig. 1. In the experiment two equally polarized beams of a pulse-repetitive Nd:YAG laser at $\lambda = 1064$ nm were used as pump radiation to create LIGs in propane/nitrogen and propane/air mixtures. The Nd:YAG laser provided pulses of 30 mJ with a duration of 7 ns, which were splitted into two pump beams. The pump and the probe beams were focussed by a lens ($f = 1000$ mm) and crossed at the appropriate angles. The crossing angle of the pump beams was 2.04° providing the fringe spacing of about 30 µm. The dimensions of the probe volume are estimated to be 300 µm in diameter and 10 mm in length. The temporal evolution of the gratings has been recorded by diffracting a cw Ar^+-laser beam (514.5 nm, 70 mW). Signal detection was accomplished using a photomultiplier tube in combination with an optical fiber, acting as a spatial filter, and an interference filter centered at the probe wavelength. The photomultiplier signal was supplied to a 500-MHz digital oscilloscope, which was connected to a computer.
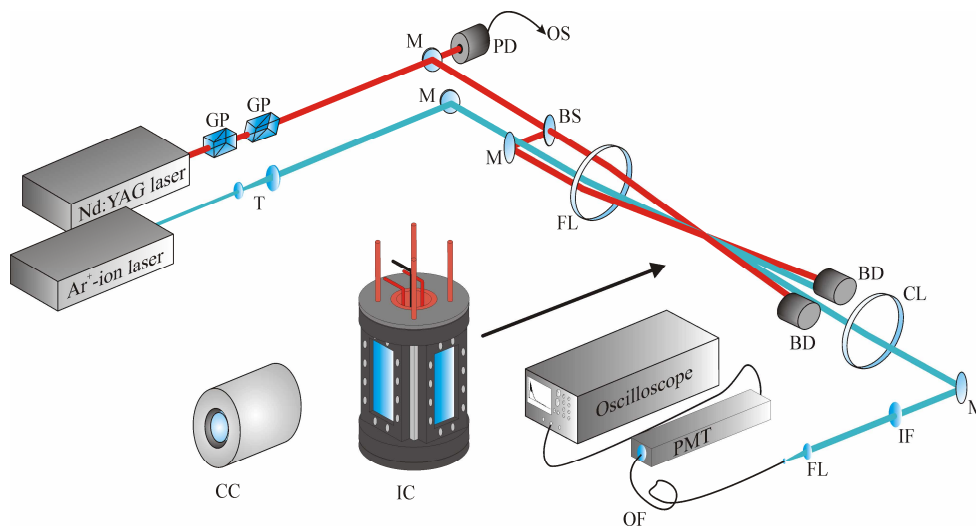

Fig. 1. Scheme of the experimental setup. (GP: Glan polarizing prism, T: telescope, BS: beam splitter, PD: trigger photo diode, OS: digital oscilloscope, M: mirror, FL: focusing lens, IC: injection chamber, CC: heatable calibration cell, BD: beam dump, CL: collimating lens, OF: optical fiber, PMT: photomultiplier tube; IF: interference filter).

With the developed setup, concentration measurements in propane/nitrogen and propane/air mixtures have been performed in two different pressurized environments: (1) in a heatable cell under stationary conditions - for calibration purposes; (2) in a non-reacting jet from a gasoline direct injection (GDI) system fueled with gaseous C_3H_8. The injection chamber has an inner diameter of 100 mm and a height of 200 mm. It is equipped with three rectangular borosilicate glass windows (Schott BK7) providing the optical access to the probe volume inside the chamber. A high-pressure prototype GDI injector with a nominal cone angle of 60° is used. The duration of the injector driving pulses, that were synchronized with

the pump laser shots, could be varied in the range of a few ms. The LIG probe volume was positioned at a distance of 45 mm downstream the injector nozzle orifice at the axis of the injection cell. The chamber pressure and temperature were maintained at about 3 bar and 345 K, respectively. To ensure that fuel of a previous injection cycle is removed from the chamber before the next injection starts, a continuous air flow through the chamber was provided with a velocity of 0.2 m/s. The injection rate was kept constant at 1 Hz throughout the experiments to additionally ensure that the previously injected fuel is taken out of the chamber before a new measurement occurs. Further details about the injection chamber and its operation can be found elsewhere [22].

3. Calibration measurements and data evaluation

Propane molecules weakly absorb radiation at $\lambda = 1.06$ μm ($\alpha \approx 10^{-4}$ cm^{-1} bar^{-1} at ambient temperature) within overtone or combination bands. The excitation of C_3H_8 molecules is followed by rapid relaxation of their internal energy due to collisions. As a result, two different LIG formation mechanisms are observed in gas mixtures with small concentration of propane: electrostriction and thermalization. The amplitude of the electrostrictive contribution is proportional to the overall gas pressure, while that of the thermal contribution - to the partial pressure of the absorbing propane molecules. Due to interference the thermal contribution may significantly modify the profile of the observed LIG signals and enhance their strength. Therefore, depending on the C_3H_8 concentration or mole fraction, x (given in %), two different evaluation procedures were applied.

For $x \leq 1.6$ % it was found to be possible to determine concentrations from the ratio $R = I_1/I_2$ of the amplitudes I_1 and I_2 of the first two oscillation peaks of the LIG signals with clearly defined interfering electrostrictive and thermal contributions, as displayed in Fig. 2(a) for a cell measurement. The single-shot LIG signal profile of Fig. 2(a) was recorded in an $x \approx 0.8$ % propane/nitrogen mixture at $p = 2$ bar and $T = 335$ K. The LIG signal peaks amplitudes ratio R is defined by the ratio of the amplitudes of thermal and electrostrictive contributions, which is proportional to C_3H_8 mole fraction x. Calibration measurements of the dependence of the ratio R on C_3H_8 concentration in the range $x = 0.2$-1.6 % were performed in the cell under stationary conditions. The calibration data were obtained in the pressure range of 1-4 bar and at cell temperature of 298 K. It was found that even though the LIG signal profile and strength are changing with pressure increase, the measured value of R is practically not dependent on pressure in the range under investigation. This can be confirmed by theoretical considerations, similar to those outlined in [23], if one assumes that the instantaneous relaxation is the main reason for the thermal contribution.

Following the same theoretical approach one can also show that the ratio of the thermal and electrostrictive contributions differs by only about 3 % in nitrogen and air. Hence, the calibration curve obtained for propane in nitrogen can be also used to define propane concentrations in propane/air mixtures. In addition, it should be noted that under the assumptions mentioned above the ratio of the thermal and electrostrictive contributions scales as $T^{-1/2}$. Since during the injection process under study temperature in the cell does not exceed 345 K, the ratio of these contributions should change by less than 8 %, as related to that at the calibration temperature. Therefore, in the first approximation the calibration dependence measured at room temperature can be regarded as applicable for the characterization of propane concentrations during the injection process without corrections for gas temperature.

The results of calibration measurements based on 100 accumulated single-shot signals are displayed in Fig. 2(b). The temperature in the cell was 298 K, and the lowest detected propane concentration was $x = 0.2$ %. The solid line in Fig. 2(b) is the fitted exponential curve with the indicated fitting parameters.
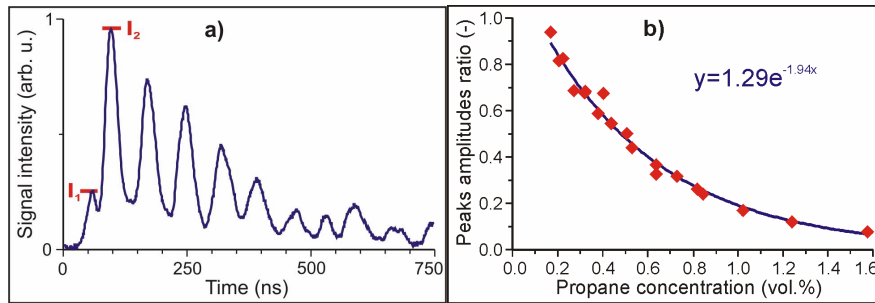
Fig. 2. (a) Measured single-shot LIG signal with clearly defined interfering electrostrictive and thermal contributions; I_1 and I_2 are the amplitudes of the first two oscillation peaks employed to derive the propane concentration; ($p_{cell} = 2$ bar; $T_{cell} = 335$ K). (b) Calibration data used to derive propane concentrations in the range of 0.2-1.6 %; (pressure range: 1-4 bar; $T_{cell} = 298$ K).

The distribution of C_3H_8 concentrations, evaluated using the peaks amplitudes ratio, from 100 single-shot signals recorded under stationary conditions in the cell with nitrogen ($p_{cell} = 2$ bar, $T_{cell} = 335$ K), containing propane at known concentration of about 0.8 %, appeared to be nearly Gaussian. The mean value obtained is $x = 0.78$ %, while the relative standard deviation, that can be taken as an estimate of the measurement error, is $\sigma_R = 0.14$.

For $x \geq 1.6$ % the thermal contributions to the LIG signals are strongly dominating, and therefore the concentration can no longer be determined from the ratio of the peaks amplitudes. In this case the information on x can be derived from the signal oscillation period, T_g. As an example, Fig. 3(a) shows a single-shot LIG signal profile recorded in a 6 % of propane in air mixture ($p_{cell} = 2.7$ bar, $T_{cell} = 345$ K), where only the thermal contribution is directly observed. The oscillation period was defined to be $T_g = 79.5 \pm 1.4$ ns. In Fig. 3(b) the concentration dependence of T_g is shown, calculated for C_3H_8 concentrations up to 10 %, using the relation describing the dependence of a binary mixture adiabatic sound velocity on species concentration and the known values of the appropriate gas parameters. One can readily show that at small C_3H_8 concentrations $T_g \approx T_g^0 \cdot (1 + 0.56 \cdot x)$.

The single-shot data allow to estimate the relative standard deviation of concentrations derived from the oscillation periods (i.e. the measurement error) as $\sigma_T \approx 0.018 \cdot T_g^0 / (T_g - T_g^0)$. In particular, $\sigma_T \approx 0.39$, if the concentration is derived using the signal in Fig. 3(a). Obviously, σ_T reduces when T_g increases with x. By assuming $\sigma_T \leq 1$ one can get an estimate of the lower limit of C_3H_8 concentration that can be defined with reasonable accuracy, if this approach is employed: $x_{min} \geq 3.2$ %. For the range of the intermediate C_3H_8 concentrations, i. e. $x \approx 1.6$-3.2 %, the procedure of signal profile fitting, with x as the fitting parameter, can be used. It takes into account variations of the ratio of electrostrictive and thermal contributions - in frame of a certain relaxation model, as well as that of the oscillation period and peaks position (oscillation phase shift), with propane concentration. However, this procedure is more elaborate and time-consuming in data treatment.
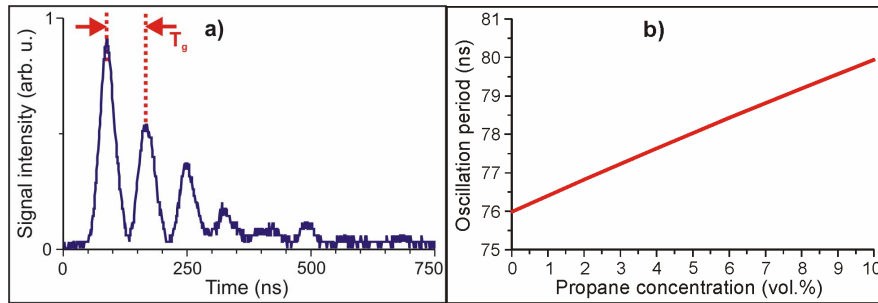
Fig. 3. (a) Thermal contribution dominated single-shot LIG signal recorded in a 6 % of propane in air mixture (p_{cell} = 2.7 bar, T_{cell} = 345 K); the oscillation period T_g = 79.5 ns is evaluated from the signal profile. (b) Calculated propane concentration dependence of T_g used to define concentrations in the range of 1.6-10 %.

4. Experimental results

The constant volume injection chamber experiments have been conducted in order to follow the temporal variation of the C_3H_8 concentration, and consequently that of the equivalence ratio, in the probe volume. Propane heated up to 330 K was injected at pressure of 13 bar. The injector driving pulse length was set at 5 ms. The chamber pressure and temperature were stabilized at 3 bar and 345 K, respectively. When gaseous propane is injected and measurements are taken at increasing delays after the start of the injector driving pulse, a sequence of values as presented in Fig. 4 can be derived from the recorded LIG signals. The delays have been increased in steps varying between 0.2 and 1 ms. The points represent the mean of up to 25 values, each derived from a single-shot measurement. A steep increase of the C_3H_8 concentration can be seen at 1.1-1.2 ms delay, when the injected gas reaches the probe volume. During about 4.5 ms a constant C_3H_8 concentration x = 7.0 %, corresponding to the equivalence ratio of about 1.8, could be derived. At larger delays the C_3H_8 concentration decreases again as the fuel cloud is moving away from the probe volume. At delays as large as, e. g., 10 ms the evaluated equivalence ratio reduces to approximately 0.1.

At the injection pulse front edge and large delays, when propane concentrations are lower than 1.6 %, the equivalence ratios were determined from the peaks amplitudes ratio of the signal profiles with the interfering electrostrictive and thermal contributions. These values are indicated in Fig. 4 by red squares. As an example, the error bars corresponding to a standard deviation at stationary conditions, are shown with one of the points. The evaluated relative standard deviation of 100 single-shot measurements at, e. g., 10 ms delay is $\sigma^i_R \approx 0.30$, which is higher than the estimated σ_R value - probably, because of pulse-to-pulse fluctuations of C_3H_8 propagation in the injection chamber.

At delays when C_3H_8 concentrations are higher, the signal oscillation period was used to derive the equivalence ratios. The corresponding values are indicated in Fig. 4 by blue circles, with the standard deviation for stationary conditions, shown as the error bars with one of these points. The relative standard deviation of 100 single-shot measurements at, e. g., 4 ms delay is $\sigma^i_T \approx 0.49$. One can consider the defined pairs of values σ_T, σ^i_T and σ_R, σ^i_R to be in a reasonable agreement with each other, since they provide the comparable, at different delays, estimate of the relative standard deviation of propane concentration fluctuations in the probe volume from one injection pulse to another ($\sigma^i \approx 0.28$).
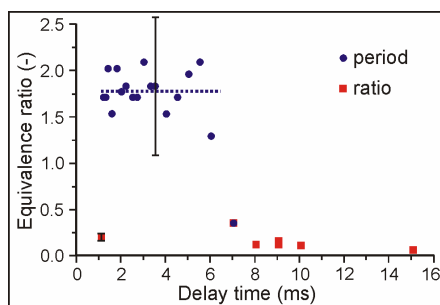
Fig. 4. Temporal behavior of the local equivalence ratio during the injection of gaseous propane (red squares: evaluation using the peaks amplitudes ratio; blue circles: evaluation using the oscillation period; dotted line - the level of the average equivalence ratio during the injection pulse). The error bars correspond to the standard deviation of measurements at stationary conditions.

5. Summary

The measurement of the temporal evolution of the local equivalence ratio by use of laser-induced gratings has been successfully demonstrated. As an example, a non-stationary gas injection process was investigated, and experimental data derived from single-shot measurements are presented. The characteristic parameters of LIG signal profiles - like the ratio of the oscillation peaks amplitudes and the oscillation period - have been employed to derive propane concentrations in the range of 0.2-10 % and to calculate the corresponding mixture equivalence ratios. Depending on the evaluation procedure, the respective relative uncertainties, which have been determined from additional cell measurements, are estimated to be in the range of 14-39 %. Since the LIGs technique shows the potential towards temporally and spatially resolved measurements of the mixture equivalence ratio, its applicability to gas phase investigation of in-cylinder applications within internal combustion engines prior to ignition will be of interest for future studies.

Acknowledgment