

Quantitative measurements of OH concentration fields by two-dimensional laser-induced fluorescence

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Abstract. Two-dimensional laser-induced fluorescence measurements (2-D LIF) of the hydroxyl radical distribution in a laminar, premixed methane/air flat flame at various pressures were performed. Instantaneous and time averaged concentration fields were determined for three different pressures (1, 5, 20 bar). The 2-D LIF data were set to an absolute scale by a calibration with a novel, single pulse one-dimensional UV absorption measurement made simultaneously with only one intensified CCD camera. To compensate effects of beam steering a spherical lens was inserted into the sheet path after the flame. The measurements are compared with results from numerical calculations.

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Combustion processes are characterized by the complex interaction between different transport processes and numerous chemical reactions. To study the various processes and to validate numerical calculations, non-intrusive in-situ measurements of concentration and temperature are necessary. For this purpose several laser-based spectroscopic methods have been developed and successfully applied in recent years [1–3]. As most technical combustion systems are turbulent, multidimensional single-pulse diagnostics is required. Two-dimensional laser-induced fluorescence (2-D LIF) is a powerful technique to measure many of the chemically important species, and temperature in a flame, with high spatial and temporal resolution with single pulse capability [4].

For 2-D LIF a laser beam is formed into a light sheet by several lenses. The light sheet intersects the flow field in a two-dimensional plane and excites the molecules or atoms. The occurring fluorescence is collected with a two-dimensional detector [5, 6] such as an image intensified CCD camera [7].

Concentration fields of OH radicals are of special interest owing to its important role in combustion chemistry and for comparison with results from reduced reaction schemes. Although qualitative results are sometimes valuable, often quantitative data are necessary. However, quantification is not a trivial problem, especially at higher pressures. Usually other

spectroscopic methods, e.g. absorption spectroscopy [8–12] have to be used to calibrate LIF data.

In the present paper we propose to combine planar LIF measurements with absorption measurements at any height above the burner surface (1-D absorption) on a single pulse basis. The calibration of the LIF signals at any axis in the plane by a separate determination of the absorption along that axis reduces strongly the influence of an unknown quenching rate, since absorption measurements are less influenced by quenching. One could consider the proposed method to be just a 1-D absorption measurement. However, the LIF measurements are needed not only to determine the length of the absorption axis but also the distribution of the OH radicals along the absorption path.

1 High pressure burner

The flame under study was a methane/air flat flame stabilized on a water cooled sinter plate of 20 mm diameter. This burner, constructed following drawings of O.N.E.R.A. allows steady combustion within a pressure range between 1 and 40 bar. Gas velocity and mixture composition were controlled by mass flow controllers. Thereby mixing ratios λ (air to fuel ratio) between 0.5 and 1.3 are possible though most measurements were done for $\lambda = 1$ and $\lambda = 1.1$. Optical access is realized by four suprasil windows orthogonal to each other. Some major advantages of this type of burner are the very high stability of the flame, which allows time integrated measurements, and the fact that similar devices are available at several other groups like O.N.E.R.A. and DLR Stuttgart. So data of these research groups can be compared directly.

2 Laser and detection system

To detect OH radicals using 2-D LIF several detection schemes have been published. Advantages and disadvantages of these schemes are discussed by Ketterle et al. [13]. The absorption of the (3–0) band in the $A^2\Sigma-X^2\Pi$ system [14] is too weak to be measured with pulsed laser systems. Excitation

of the (0–0) band with detection in the (0–1) band at 343 nm cancels out due to the low signal level. Higher laser power results quickly in saturation. The (1–0) excitation was preferred to the (0–0) excitation with detection in the (0–0) band due to reduced problems with fluorescence trapping [15] and stray light.

The OH radicals were excited using the $P_1(8)$ line of the (1–0) vibrational band of the electronic $A^2\Sigma X^2\Pi$ transition. The thermal population of the initial $N'' = 8$ state varies only by 10% between 1300 K and 3200 K and can therefore be considered as independent of temperature [3]. Figure 1 shows the experimental set-up. The output of a frequency doubled, YAG pumped dye laser (Quantel YG 580/TDL 50) was formed into a light sheet (13 mm \times 0.5 mm) using cylindrical lenses. Optical elements and diaphragms reduced the laser energy from 3 mJ at the laser output to 1 mJ in the flame. The laser linewidth determined from OH lines in a low pressure discharge has been 0.16 cm^{-1} , well below the 0.4 cm^{-1} of the observed OH absorption lines in the flame at 1 bar but large enough to cover the Doppler width. This, together with the unusual long pulse duration (10.0 ± 0.3 ns) of our laser system greatly reduced problems with saturation of the OH transition compared to other YAG-based systems. Emission of the excited OH molecules was detected in the (0–0) and (1–1) band around 314 nm with an intensified, slow-scan CCD-camera (Princeton Instruments, 14 bit) which was oriented perpendicular to the light sheet. The camera was operated in the gated mode with 120 ns gate time to suppress flame emission. Synchronization between excitation and detection system was checked with a fast digital storage oscilloscope. The fluorescence light was collected using a standard UV optics with a focal length of 105 mm and $f_{\#} = 4.5$ (Nikon, UV-Nikkor). The spatial resolution of the detection system within the plane of detection has been about 100 μm . A UV cutoff filter (WG 295) was used to suppress Rayleigh scattering, straylight signals from quartz and metal surfaces, and the OH (1–0) emission. Since measurements near the burner surface were desired, a background subtraction was necessary to eliminate reflections of the light sheet on the sinter plate.

To determine the absorption the incoming and outgoing light sheet intensities have to be measured. For that purpose the light sheet profiles before and after the burner were imaged

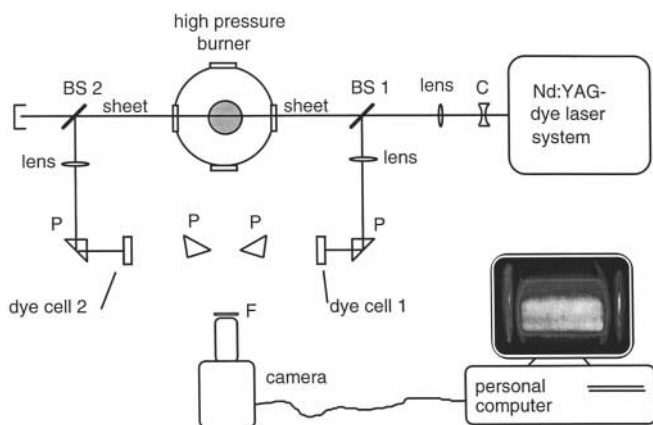


Fig. 1. Experimental arrangement for 2-D LIF measurements combined with 1-D UV absorption spectroscopy of OH radicals (BS: 5% beam splitter, P: prism, F: filter, C: cylindrical lens)

onto a quartz cell containing a fluorescent dye. The UV light intensity distributions are converted into visible light and were imaged via prisms onto the same CCD detector as the OH signal (Fig. 1). It was important to make sure that these three images were at the same distance to the plane of the detector.

At pressures exceeding 5 bar beam steering becomes significant (Fig. 2a) due to fluctuations of the index of refraction at the edges of the flame (“Schlieren” effects) [16]. For pressures higher than 20 bar the shape of the light sheet was totally distorted after passing the flame (Fig. 2b). Figures 2a and 2b show single pulse measurements of the intensity distribution of the light sheet after passing the flame at a distance of 720 mm.

Evaluation of such data is not possible as it is not known which part of the outgoing light sheet corresponds to a given part of the incoming light sheet. To restore the shape of the light sheet we placed a simple spherical lens ($f = 180$ mm) at a distance of $2f$ to the burner axis and $2f$ to the dye cell (Fig. 2c, d). A similar set-up was proposed by Kohse-Höinghaus et al. [17] to reduce the motion of a laser beam after passing the flame of a high pressure burner.

For a quantitative evaluation of the data several checks for linearity were necessary. The linearity of the following points was verified: Conversion of the UV laser beam to the visible light in the dye cell, signal conversion on the MCP-CCD detector and homogeneity of the camera system. Since homogeneity of the camera system was not perfect, we corrected the fluorescence images for pixel-to-pixel variations in sensitivity [18]. To check the linearity of the LIF signal with respect to saturation measurements were performed for different laser energies. While the presented images were taken for pulse energies of 1 mJ, we also performed measurements for energies down to 100 μJ . Despite the increasing noise level the results were identical for each laser energy within the estimated errors. This corresponds to given saturation parameters in [19–21].

3 Data acquisition and image processing

A typical image recorded by the CCD camera is depicted in Fig. 3. It contains three parts: the incoming light sheet profile on the right side, the outgoing light sheet profile on the left side, and the raw OH-LIF signal in the centre. As the LIF signal is a function of the laser intensity and its profile, this raw image is not yet a direct measure of the OH concentration.

The recorded intensity distributions of the light sheet profiles contain the information of the integral absorption along lines parallel to the burner surface. To extract the integral absorption data two images have to be recorded, one with the laser tuned to a molecular absorption (resonant case) and one with the laser detuned (nonresonant case). In the latter case indeed no fluorescence was detected. This is not necessarily the case with fuel rich flames due to the broad band absorption and fluorescence of large transient hydrocarbons.

The two-dimensional images of each light sheet profile were transferred into one-dimensional intensity distributions by calculating a mean value for each line, i.e. for each height above the burner surface. Not only in the resonant case but also in the nonresonant case the incoming and the outgoing intensity distributions were not identical. While in the resonant case the differences are expected due to absorption, in the nonresonant case differences persist due to dust and distortion of

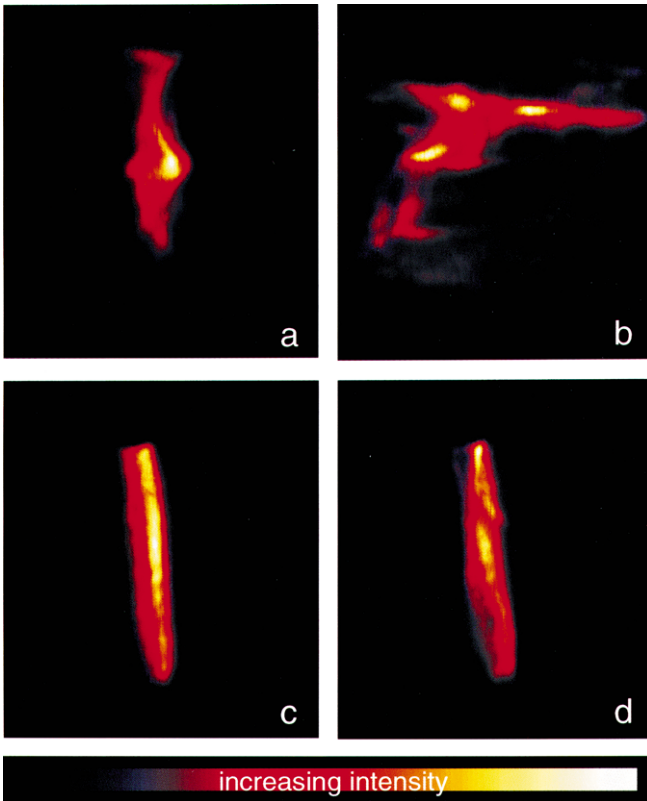


Fig. 2a–d. Intensity distribution of the light sheet after passing the flame at a distance of 720 mm. While in (a) at 10 bar the shape of the light sheet is still recognizable, in (b) at 20 bar the shape of the light sheet is totally distorted. A spherical lens is able to reproduce the shape of the light sheet, which is shown in (c) (10 bar) and (d) (20 bar)

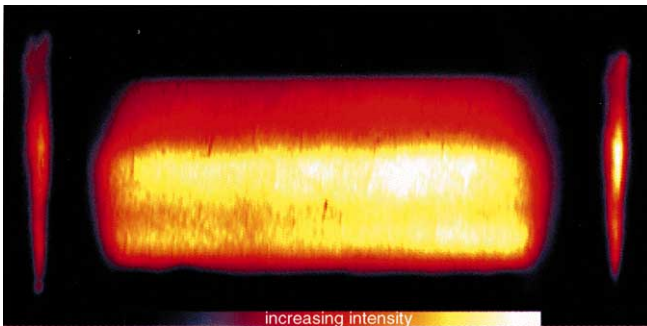


Fig. 3. Single pulse measurement of the OH fluorescence (raw data) including the incoming (right side) and outgoing light sheet profile (left side) at $p = 1$ bar and $\lambda = 1.0$

the optical elements. Inserting a mesh into the incoming light sheet generates a pattern which allows the precise determination of which part of the incoming light sheet corresponds to a given part of the outgoing light sheet. With this information, using the non-resonant light sheet profiles, we can determine a transform function which describes the imaging of the incoming light sheet on the outgoing light sheet. This transform function is only determined by the optical setup and will not change during the measurements. Dividing the transformed intensity profiles of outgoing and incoming light sheet results in the corrected transmission T of the laser light as a function of height above the burner. Using the non-resonant data one

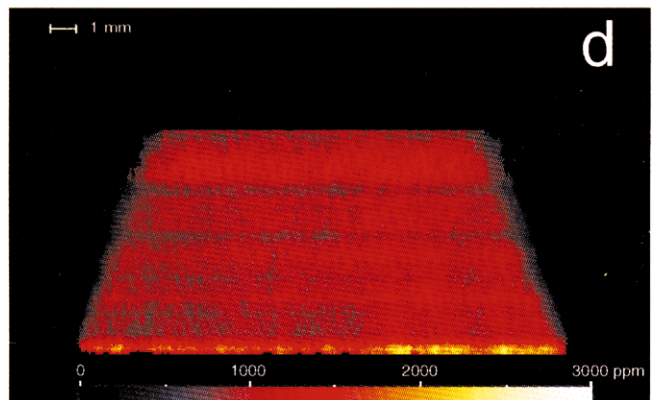
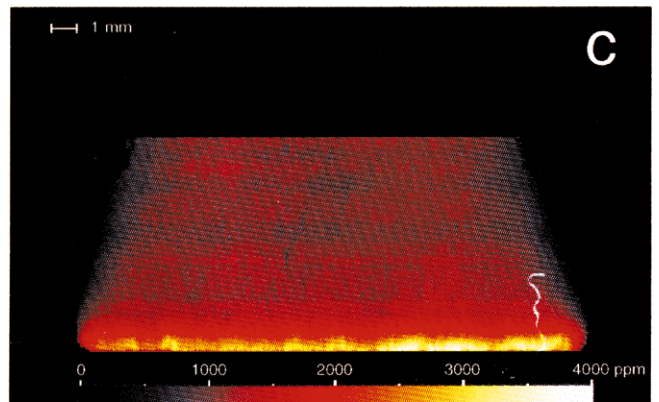
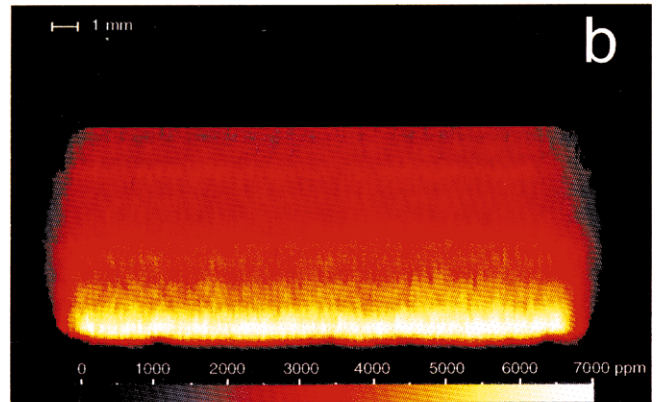
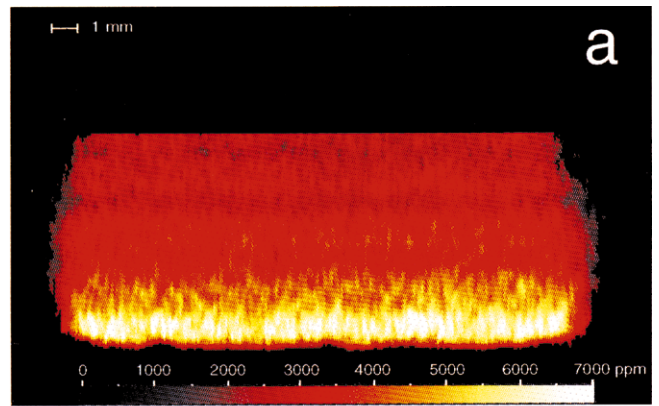


Fig. 4a–d. Two-dimensional absolute OH radical concentration fields. **a** Single pulse measurement at $p = 1$ bar, $\lambda = 1.0$; **b** averaged over 10 laser pulses at $p = 1$ bar, $\lambda = 1.0$; **c** averaged over 100 laser pulses at $p = 5$ bar, $\lambda = 1.1$; **d** averaged over 100 laser pulses at $p = 20$ bar, $\lambda = 1.1$

gets unity for each height. Using the resonant data one gets the percentage of the light which was not absorbed by the OH radicals in the flame. So the part of the light absorbed by the OH radicals is given by $1 - T$.

The evaluation of the absolute concentration (following the procedure given in [22]) using Bouguer–Beer's law needs, however, some additional data. The absorption length can be determined from the 2-D LIF image taking into account the magnification factor of the imaging system. The absorption coefficient can be calculated using Einstein coefficients for absorption and line positions listed in the literature [23,24]. The partition function was calculated by summing up Boltzmann factors of all molecular levels which are significantly populated at flame temperature. The absorption line shape was measured as a function of pressure by careful scanning of the $P_1(8)$ line of the (1–0) band. This line is not affected by neighbouring lines even at pressures up to 20 bar [21]. A Voigt profile turned out to be best suited to fit the line shape data. Temperature data were provided by CARS measurements [25,26] and by numerical simulation. Since the thermal population of the $N'' = 8$ state varies only slightly in the temperature range of interest, minor errors in the temperature data have no serious consequences.

Due to the mutual proportionality of concentration, absorption, and fluorescence the fluorescence image can be used to distribute the integral concentration values to different locations along the direction of light propagation. We used the iterative algorithm for one-dimensional LIF data of Hertz et al. [11] line by line, i.e. for each height above the burner surface to get the two-dimensional OH concentration fields.

4 Results and discussion

Figure 4a shows an absolute concentration field in a steady methane/air flame ($\lambda = 1.0$, $p = 1$ bar). This image was taken with a single laser pulse. The signal-to-noise ratio (SNR) is limited by the number of photons detected per pixel. This was demonstrated by averaging over several laser pulses which considerably increased the quality of the 2-D LIF images (Fig. 4b). Even at higher pressures concentration fields were determined with good SNR as shown in Figs. 4c and 4d for pressures of 5 and 20 bar.

OH radicals are formed in the flame front and are also present in the hot burnt gases according to the thermal equilibrium concentration. The flame front can be seen clearly in Figs. 4a–d as a bright zone in the lower part of the images. Increasing pressure results in a decreasing reaction zone thickness. It can be seen from the noise level that the signal level is decreasing for increasing pressure. Although increasing pressure results in an increasing absolute number density of OH radicals, the signal level decreases due to mole fraction decrease, absorption line width broadening, and increase of electronic quenching.

Since calibration of the 2-D LIF data is performed by 1-D absorption measurements the influence of quenching is reduced. In addition it was shown (see e.g. [27–30]) that changes in quenching rate due to stoichiometry and position in the flame are small in burnt-gas regions of premixed hydrocarbon/air flames. Larger effects are only observable in the flame front. Since in our special case changes in species

concentration and temperature are mainly normal to the flame front and the calibration is performed for each height parallel to the flame front, signal changes due to quenching rate variations in this direction are negligible.

Changes in the quenching rate as a function of the position in the flame are dependent on the stoichiometry. It was shown [20,29] that these changes are lower for lean flames than for fuel rich flames. In a premixed ethylene/air flame with $\lambda = 1.07$ Schäfer et al. [20] determined variations in quenching rate to be less than 5% whereas for $\lambda = 0.61$ differences of about 20% were observed. Therefore quenching corrections

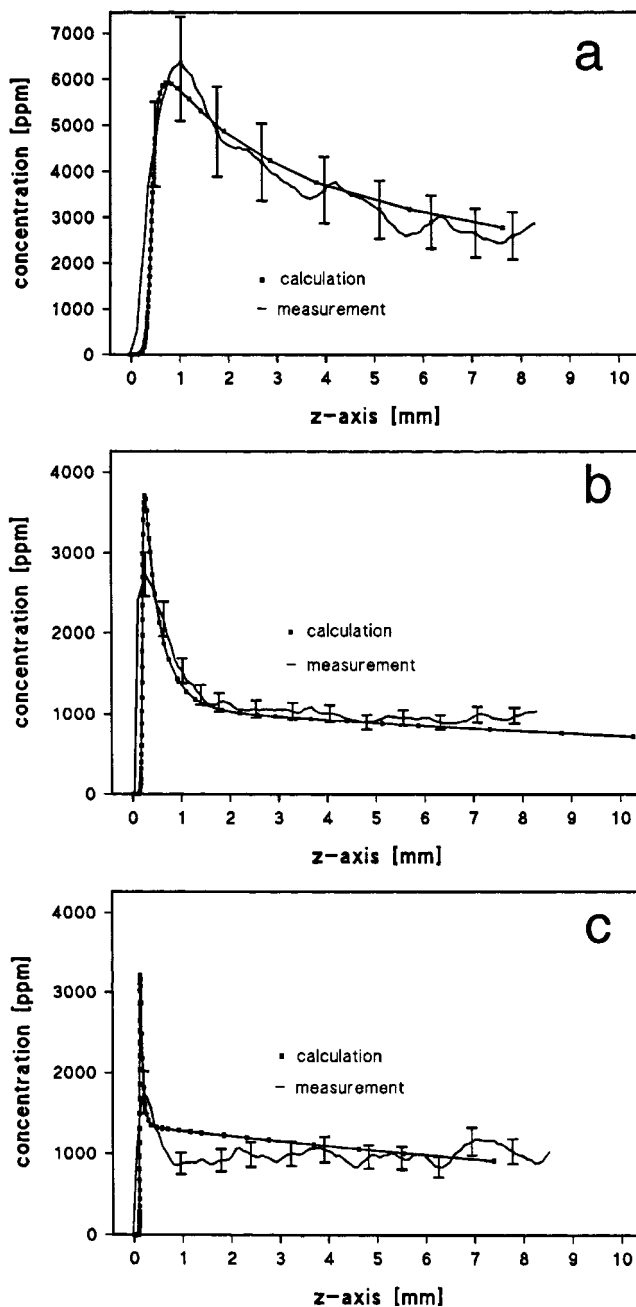


Fig. 5a–c. OH radical concentration profiles; comparison between measurements including estimated errors and calculation. Profiles normal to flame front, (a) single pulse measurement at $p = 1$ bar, $\lambda = 1.0$; b averaged over 100 laser pulses at $p = 5$ bar, $\lambda = 1.1$; c averaged over 100 laser pulses at $p = 20$ bar, $\lambda = 1.1$.

are of minor importance because our aim is the application of 2-D LIF in industrial combustion systems which tend to extreme lean flames.

The results are subject to several limitations and errors like shot noise, inaccuracy of spectroscopic data, absorption length, transmission measurement, and temperature. The combination of these types of errors leads to an estimated total accuracy of 20% for single pulse images and 10% for averages over 10 pulses at 1 bar. For higher pressures, the decreasing signal level leads to an increasing error. By repeating the measurements the OH concentration profiles were reproduced within 10% for the averaged data at 5 bar and 15% at 20 bar.

Although fluorescence trapping with the used excitation scheme is less severe than with excitation of the (0–0) band with detection in the (0–0) band, it cannot be neglected. Using Franck–Condon factors [31], VET, and quenching data [30] fluorescence trapping for the longest possible path across the flame was estimated to be smaller than 10%. However, the measurements indicate a considerable lower effect. Assuming a homogeneous distribution of OH radicals in planes parallel to the burner surface, fluorescence light starting from the middle of the flame should be noticeably diminished compared to fluorescence emitted at the edge of the flame. However, there is no evidence in the fluorescence images.

Since a major challenge in the application of laser diagnostics in combustion is to provide quantitative data for comparison with numerical simulation models, a comparison for different pressures is shown in Figs. 5a–c. The figures show a typical profile through the measured OH concentration fields normal to the flame front and the results of a one-dimensional calculation based on the one-dimensional flat flame burner code [32]. Figures 5b and 5c result for averages over 100 laser pulses whereas Fig. 5a has been taken from single pulse data. We have not succeeded yet stabilizing the flame front height at elevated pressures. The flame front moves to and fro and appears in the averaged images shown in Figs. 5b and 5c to be broader than it actually is.

In conclusion, we have established a two-dimensional measuring technique for determination of absolute OH concentration fields on a single laser pulse basis. Briefly, the method is a combination of 2-D LIF and 1-D absorption spectroscopy. An application up to 20 bar was demonstrated. Comparison with numerical model calculations showed very good agreement.

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References

1. A. Eckbreth: *Laser Diagnostics for Combustion Temperature and Species* (Abacus Press, Tunbridge Wells 1988)
2. K. Kohse-Höinghaus: *Prog. Energy Comb. Sci.* **20**, 203 (1994)
3. A. Arnold, H. Becker, R. Hemberger, W. Hentschel, W. Ketterle, M. Köllner, W. Meienburg, P. Monkhouse, H. Neckel, M. Schäfer, K.P. Schindler, V. Sick, R. Suntz, J. Wolfrum: *Appl. Opt.* **29**, 4860 (1990)
4. R.K. Hanson, J.M. Seitzman, P.H. Paul: *Appl. Phys. B* **50**, 441 (1990)
5. M.J. Dyer, D.R. Crosley: *Opt. Lett.* **7**, 382 (1982)
6. G. Kychakoff, R.D. Howe, R.K. Hanson, J.C. McDaniel: *Appl. Opt.* **21**, 3225 (1982)
7. R. Suntz, H. Becker, P. Monkhouse, J. Wolfrum: *Appl. Phys. B* **47**, 179 (1988)
8. V. Sick, A. Arnold, E. Diebel, T. Dreier, W. Ketterle, B. Lange, J. Wolfrum, W. Thiele: in *Twenty-third Symposium (International) on Combustion* (The Combustion Institute, Pittsburgh 1990) p.495
9. J.H. Bechtel, R.E. Teets: *Appl. Opt.* **18**, 4138 (1979)
10. W.R. Anderson, L.J. Decker, A.J. Kotlar: *Combust. Flame* **48**, 179 (1982)
11. H.M. Hertz, M. Aldén: *Appl. Phys. B* **42**, 97 (1987)
12. E.W. Kaiser, K. Marko, D. Klick, L. Rimai, C.C. Wang, B. Shirinzadeh, D. Zhou: *Combust. Sci. Technol.* **50**, 163 (1986)
13. W. Ketterle, M. Schäfer, A. Arnold, J. Wolfrum: *Appl. Phys. B* **54**, 109 (1992)
14. P. Andresen, A. Bath, W. Gröger, H.W. Lülff, G. Meijer, J.J. ter Meulen: *Appl. Opt.* **27**, 365 (1990)
15. A. Lawitzki, R. Tirgrath, U. Meier, K. Kohse-Höinghaus, A. Jörg, T. Just: in *Proceedings of the Joint Meeting of the German and Italian Sections of the Combustion Institute*, Ravello, Italy (1989) paper 1.4
16. F. Weinberg: in *Twenty-sixth Symposium (International) on Combustion* (The Combustion Institute, Pittsburgh 1996) in press
17. K. Kohse-Höinghaus, U. Meier, B. Attal-Trétout: *Appl. Opt.* **29**, 1560 (1990)
18. G. Kychakoff, R.D. Howe, R.K. Hanson, K. Knapp: AIAA reprint 83-0405, 21th Aerospace Sciences Meeting, (Reno 1986)
19. D. Klick, E.W. Kaiser: *Appl. Opt.* **23**, 4184 (1984)
20. M. Schäfer, W. Ketterle, J. Wolfrum: *Appl. Phys. B* **52**, 341 (1991)
21. B.E. Battles, R.K. Hanson: *J. Quant. Spectrosc. Radiat. Transfer* **54**, 521 (1995)
22. U. Boltendahl: Dissertation, RWTH Aachen (1974)
23. I.L. Chisdey, D.R. Crosley: *J. Quant. Spectrosc. Radiat. Transfer* **23**, 187 (1980)
24. G.H. Dieke, H.M. Crosswhite: *J. Quant. Spectrosc. Radiat. Transfer* **2**, 97 (1962)
25. V. Bergmann: Dissertation, DLR Stuttgart/Universität Bielefeld (1993)
26. V. Bergmann, W. Stricker: *Appl. Phys. B* **61**, 49 (1995)
27. M. Köllner, P. Monkhouse, J. Wolfrum: *Chem. Phys. Lett.* **168**, 355 (1990)
28. M. Tsujishita, A. Hirano: *Appl. Phys. B* **62**, 255 (1996)
29. A. Dreizler, R. Taday, P. Monkhouse, J. Wolfrum: *Appl. Phys. B* **57**, 85 (1993)
30. P. Beaud, P. Radi, D. Franzke, H.-M. Frey, B. Mischler, A.P. Tzannis, T. Gerber: submitted to *J. Chem. Phys.*
31. D.R. Crosley, R.K. Lengel: *J. Quant. Spectrosc. Radiat. Transfer* **15**, 579 (1975)
32. A. Schlegel, S. Buser, P. Benz, H. Bockhorn, F. Mauss: in *Twenty-fifth Symposium (International) on Combustion* (The Combustion Institute, Pittsburgh 1994) p.1019