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# Projekt
# Quantitative laserinduzierte Fluoreszenz in der Verbrennung

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## ZUSAMMENFASSUNG

The investigation of laminar counter flow flames was continued in the reporting period. An experimental setup for one-dimensional spontaneous Raman scattering measurements was implemented. With this technique, the locally resolved temperature and major species concentrations can be simultaneously determined for a 1-D (line) measurement volume. From a set of measurements at different flame locations, 2-D fields of temperature and concentrations are derived.

Experimental results for hydrogen flames are compared with predictions from direct numerical simulations (DNS) performed at the ETHZ. General agreement is found to be good. Some details in the choice of boundary conditions for the simulations will be modified for future work.

The spatially resolved, two-dimensional information on temperature and species concentrations is also used in the data analysis of 2-D OH laser-induced fluorescence (LIF) measurements. There, the spatial distribution of LIF signal is affected by collisional quenching. Determination of correction factors requires the local temperature and collider concentrations which are now known from the Raman measurements.

An attempt to record transient flame structures, while the flame shape changes from diffusion to edge flame, met with unexpected difficulties, and a different experimental approach is under development.

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# Projektziele

The objectives of the project are: 1. To investigate flame structure and flame dynamics in an opposed jet burner. 2. To make steps in the development of laser-induced fluorescence (LIF) as a quantitative measuring technique for laboratory and commercial burners.

Specifically, counter flow flames are investigated using laser and other optical diagnostic methods. A system is considered where both a planar diffusion flame and a toroidal edge flame can be stabilized. Though the cold gas flow is laminar, strain rates can be made large enough to induce extinction of the flame on the symmetry axis. This property makes the system a useful, simple model for flame quenching and re-ignition of unsteady flamelets which occur in a certain class of turbulent flames. The structure of both diffusion and edge flames, as well as the dynamics of the transition between these shapes is studied for different fuels. The observations may be compared with the results of numerical studies performed independently in a group at the *LAV laboratory* at *ETHZ*.

Phenomena observed so far include flame extinction, re-ignition, switching between different flame structures, as well as attachment of the flow field to a wall. The physical mechanisms behind these phenomena are investigated as well as the influence of the following parameters: fuel type (methane and hydrogen), fuel dilution, boundary conditions (temperature, nozzle diameters, distance between the two opposed nozzles). Various laser-based measurement methods are employed to obtain complementary information on temperature and species concentration distributions, and flow field.
The targets for this report's period included:

- One-dimensional spontaneous Raman scattering measurements for the derivation of 2-D distributions of temperature and major species concentrations.
- Use of this 2-D information to apply corrections for fluorescence quenching to previously measured 2-D OH LIF images.
- Implementing a method for inducing the transition between diffusion and edge flame, and synchronizing single-pulse laser measurements with this transition, thus providing time-resolved information on the transient flame structure.

# Durchgeführte Arbeiten und erreichte Ergebnisse

## 1. Introduction

Axisymmetric counter flow burners provide a convenient setup to study strained laminar flames which play a role in the investigation of flame extinction and re-ignition, particularly for turbulent flames in the "flamelet" regime. It was found both in 2-D numerical simulations and experimentally that, under suitable conditions, a toroidally shaped edge flame can exist in addition to the well-known planar diffusion flame [1]. Furthermore, over a wide range of flow velocities, these two shapes can both exist for the same flow conditions (bistability). On the axis of the edge flame only mixing of fuel and air takes place, but no reaction. Fuel and oxidizer start to react at a well defined distance away from the flow axis. Under suitable conditions it is possible to “switch” between the two flame types, and a marked hysteresis is associated with this transition. This edge flame can not be found when using the 1-D codes commonly employed for the simulation of opposed flow diffusion flames.

In this reporting period, measurements of major species concentrations were made using spontaneous Raman scattering. With the experimental setup described below, spatially resolved Raman signal from a line (1-D) could be recorded simultaneously. In addition to species concentration, spatially resolved temperature information can also be derived from these measurements. Such spatially resolved information can be applied to correct 2-D OH LIF signals for the effect of collisional quenching, demonstrating again the benefits of having more than one measurement technique at one's disposal. In a separate experiment an attempt was made to obtain time-resolved OH LIF images of the transient flame, while it is in the course of quenching or re-igniting.

## 2. Experimental

### 2.1 Burner

The burner consists of two steel tubes (typical diameters: d = 2.7, 5 and 10 mm) in a vertical counter flow configuration, with water-cooled brass disks attached in the exit plane of each nozzle. The two disks form a vertical gap of width L = 1d. The length of the steel tubes is chosen such that in the exit plane a laminar parabolic flow velocity profile has developed. The atmospheric pressure burner is enclosed in a housing which protects the flame from room air draughts and excess oxygen. Four quartz windows permit access for optical diagnostics. The whole burner is mounted on a two-axis translation stage such that the flame may be displaced with respect to the measurement volume defined by the laser's focal region (which is space-fixed in laboratory coordinates). The two photographs in Fig. 1 illustrate the shapes of a diffusion and edge flame.

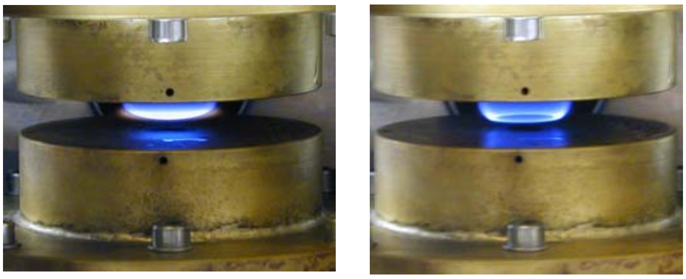

Figure 1: Methane-air counter flow flames. Left: Planar diffusion flame, with closed flame front surface on the symmetry axis. Right: Toroidal edge flame, no reaction occurs on the axis. The two flame shapes shown were obtained for identical flow conditions. The fuel flow direction is upwards, air flow downwards. Flow tube diameter d, and vertical gap size L: 10mm; disk diameter: 72mm. The laser beam traverses the flame horizontally.

### 2.2 Raman Scattering

A spontaneous Raman scattering setup for 1-D measurements was used; many components were similar to those described in [2]. A pulsed, frequency-doubled Nd:YAG laser was used. Its beam was first passed through a pulse stretcher in order to avoid the problem of dielectric breakdown in the focal region, by reducing the peak power while maintaining the overall energy in the pulse. The setup of signal collection optics, spectrograph, and detector is shown in Fig. 2. With the spectrograph dispersion and detector dimensions given in the experiment, the vibrational Raman spectrum of all major species in the flame can be detected simultaneously. Gas composition along a line (1-D) across the flame is thus analyzed in one measurement. Since all (major) species are detected, a local number density can also be derived and hence, by invoking the ideal gas law, a temperature information is obtained.

For a given flame, a series of 1-D Raman measurements was made by vertically scanning the measurement volume across the flame with a step width of 0.5 mm. The set of measurements was evaluated following the procedures discussed in [2], every single 1-D measurement providing one line profile, along a 12 mm path in the flame, for each of the major species concentrations, as well as for the local temperature. These line profiles were then combined to form a 2-D picture, and interpolated with a suitable scheme to fill in the plane. Due to stray light from the metal surfaces in the burner, a minimum distance of 1 mm had to be kept between the measurement location and the burner's disks. In the examples shown below, the tube diameter was d = 5 mm, therefore with seven 1-D Raman measurements, the distribution for a 3 mm * 12 mm region could be derived.

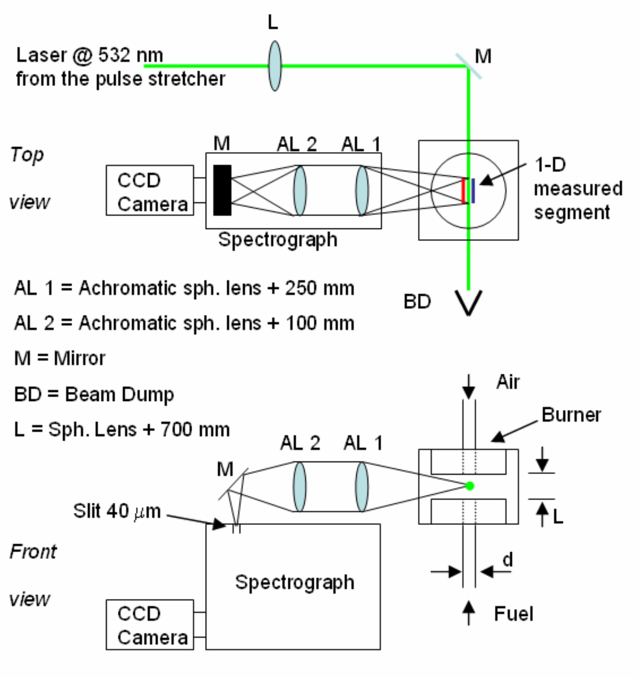

Figure 2:

Setup for 1-D Raman scattering measurements
A segment of the laser beam's focal region is imaged onto the entrance slit of the spectrograph. After spectral dispersion the signal is detected with a gated, intensified CCD camera. One axis of the detected image corresponds to wavelength (i.e. Raman shifts), the other axis to the spatial dimension (i.e. to different positions on the entrance slit, and hence in the flame).
A "notch" filter mounted in front of the entrance slit (not shown) selectively attenuates stray light at the laser wavelength.

### 2.3 Single-Shot OH LIF

One subtask calls for a method to induce the transition between the two bistable forms, diffusion and edge flame, such that a laser measurement can be synchronized with the transient event. The idea behind these measurements is to obtain single-shot images of the transient flame structure at various delay times, while the flame is in the course of quenching or re-igniting. It should then be feasible to derive the velocity of receding or progressing flame fronts, as well as the general movement of the reaction zone. These experimentally found structures may then be compared with existing DNS predictions [3] for the transients. To this end a fast, electrically switched valve (switching time < 2 msec) was implemented in the supply line for the air flow tube, which provides the possibility to increase (or reduce) the air flow velocity with a step function. The switching of this valve was synchronized with the laser pulse by means of a delay generator and custom made power supply. In this way, the transition between the flame shapes could be induced in either direction, by suitably choosing the initial fuel and air flow velocities. The apparatus used for single-shot OH LIF is identical to the one described previously [4].

## 3 Results

### 3.1 Raman Scattering

Raman scattering measurements were performed for more than 70 flames, covering a wide variety of conditions, with fuel type and composition, flow velocities, and burner nozzle diameters as the parameters. By way of example, in Figure 3 are shown results for a planar diffusion flame using 50% hydrogen (by volume) in $N_2$ as fuel, burning with air. In the left column are shown the experimentally found distributions of oxygen (a), hydrogen (c), and water (e) mole fractions; the r and z axes are normalized with the tube diameter d. The right column shows corresponding results ((b),(d)&(f)) from direct numerical simulations (DNS) performed at the ETH Zurich [5]. The 2-D simulations were carried out for the complete half plane "to the right" of the symmetry axis, the boundary conditions for the incoming fuel and air flows being imposed at height z=0 and z=1, respectively. (Note that DNS for hydrocarbon fuels is not yet a routine option, due to the increased computational effort necessitated by the much more complicated chemical reaction mechanisms of e.g. methane flames.)

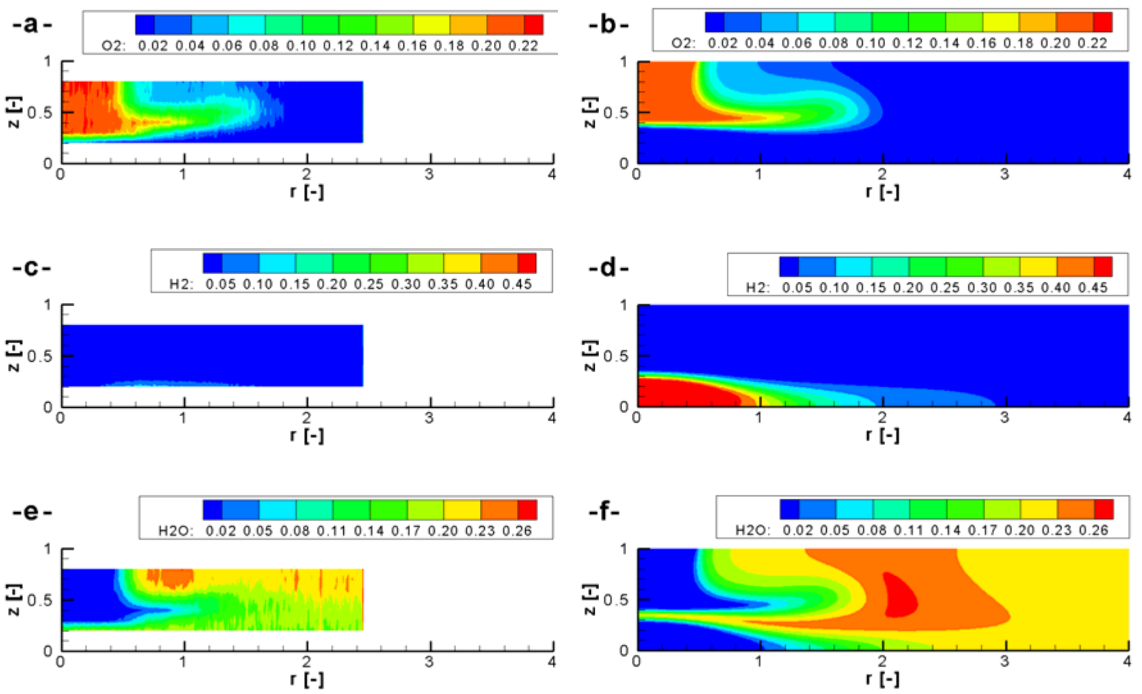

Figure 3: Comparison between experimentally found (left column) and calculated (right column) distributions of oxygen (a, b), hydrogen (c, d), and water (e, f) in a diffusion flame, using 50% hydrogen in $N_2$ as fuel. The r and z axis are scaled with tube diameter (d = 5 mm), r = 0 is on the burner symmetry axis. Fuel and air flow velocities are 3 m/s.

The results for the $O_2$ concentration show that measurements and simulations are in good agreement in the region with r>0.5, beyond the nozzle radius. Near the symmetry axis, however, the simulation predicts that $O_2$ is consumed closer to the air (upper) nozzle. The difference is even more obvious for the $H_2$ mole fraction: In the simulation $H_2$ and $O_2$ impinge at z • 0.3 - 0.4, while from the measurements it must be concluded that they meet within the first mm above the lower disk. Some $H_2O$ is present at z = 0.2, and the shape of the boundary between water and the cold air flow, with its sharply peaked extension towards larger radii at about z = 0.4, is very nicely seen in both simulation and experiment. It must be noted, however, that the locations of peak water concentrations do not agree well; in the experiment, water concentration is biased towards the upper disk.

Figure 4 compares experiment and DNS of temperature and OH distribution for the diffusion flame. The predicted temperature distribution agrees very well with the experimentally determined one. On the flow axis the experimentally seen flame front, as indicated by peak OH signals, is closer to the fuel nozzle. Also, the simulation shows, in the radial direction, a downward curvature of the OH distribution which is not present in the experiment.

Cold-flow velocity measurements [1, 6] in a similar burner configuration with disks attached have shown that the flow field itself possesses multiple asymmetric steady states, with the stagnation plane closer to either the upper or the lower nozzle. In the present case, the stagnation plane is close to the lower disk. With the stagnation plane so close to the jet exits (order-of-magnitude: 0.2 tube diameters), it is very likely that the boundary conditions imposed for the DNS have a strong effect on the results, in particular on the location and shape of the stagnation plane. In future work, the numerical domain needs to be extended "upstream" into the nozzles, such that the boundary conditions may be imposed at a position which is sufficiently far away from the stagnation point; this will provide a more accurate prediction of the flow field in the regime between the two disks.

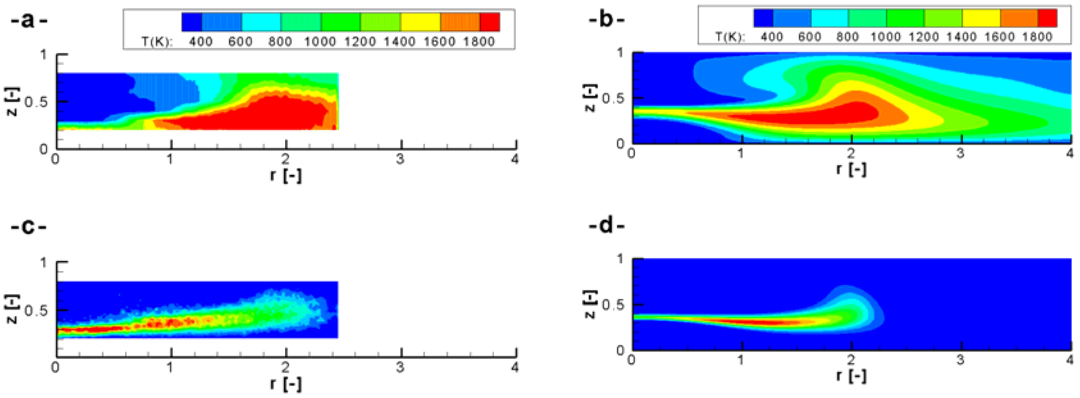

Figure 4: Comparison between experimentally found (left column) and calculated (right column) distributions of temperature (a, b) and OH radicals (c, d) in the hydrogen / air diffusion flame. Fuel and flow conditions are the same as in Fig. 3.

### 3.2 LIF Signal Corrections for Quenching

The LIF signal intensity is a function of the molecule's number density in the lower quantum state and therefore, in principle, a measure of species concentration. However, the fluorescence quantum yield and hence the LIF signal for a given transition is strongly affected by fluorescence quenching. Quenching is caused by collisions with other molecules present in the sample, and the quenching rate is a function of temperature, concentrations of the various colliders, as well as the quantum state of the excited molecule. In a flame where temperature and composition vary widely it is difficult to assess this quenching problem quantitatively. With the aid of the temperature and major species concentration fields derived from Raman scattering measurements, it is possible to correct the measured LIF signal intensities. For every pixel in the LIF image, a correction factor can be calculated by combining the (locally resolved) temperature and concentration information with the species-specific and temperature dependent quenching cross sections given in [7]. In Figure 5 is shown a comparison between the raw data of a 2-D OH LIF image recorded in an edge flame (again 50% $H_2$ in $N_2$ vs. air, 5 mm diameter burner nozzles), and the image after application of the quenching correction. Note the change in shape of the area exhibiting highest signal intensities.

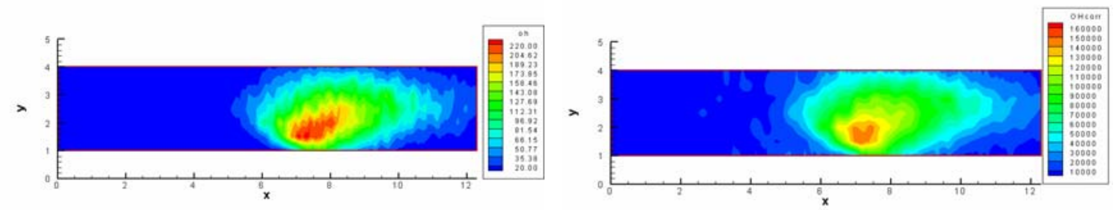

Figure 5: 2-D OH LIF image of an edge flame (50%$H_2$ in $N_2$ vs. air). Left panel shows raw data; right panel after application of quenching correction factors as described in the text. Axes are scaled in mm, x = 0 is on the burner symmetry axis (i.e. the right half plane is shown).

### 3.3 Single-Shot Measurements of Transient Flames

The valve and synchronization setup for inducing the transition between the two bistable flame shapes was implemeted, and it works as expected. However it was found that the "switching" of flame shapes itself exhibits a substantial temporal jitter (up to 10 msec) with respect to the instant when the air flow velocity was stepped up or down. The time scale of this jitter thus is larger than that of the most interesting, faster components of transient events, and the desired information on intermediate flame structures could not be obtained with sufficient accuracy and repeatability. To give an impression of the phenomena to be investigated, in Fig. 6 are shown four examples of steady-state and intermediate structures, for a transition from diffusion to edge flame, which were visualized with single-shot 2-D OH LIF. Note that because the data acquisition timing suffered from the jitter mentioned, no quantitative conclusions about the speed of the receding flame front can be drawn. The original work plan therefore has to be modified. Upon reconsidering the situation, an option currently pursued is to record a time-resolved sequence of images of one single transient event, using a camera capable of high frame rates that can be loaned from a group at ETHZ.

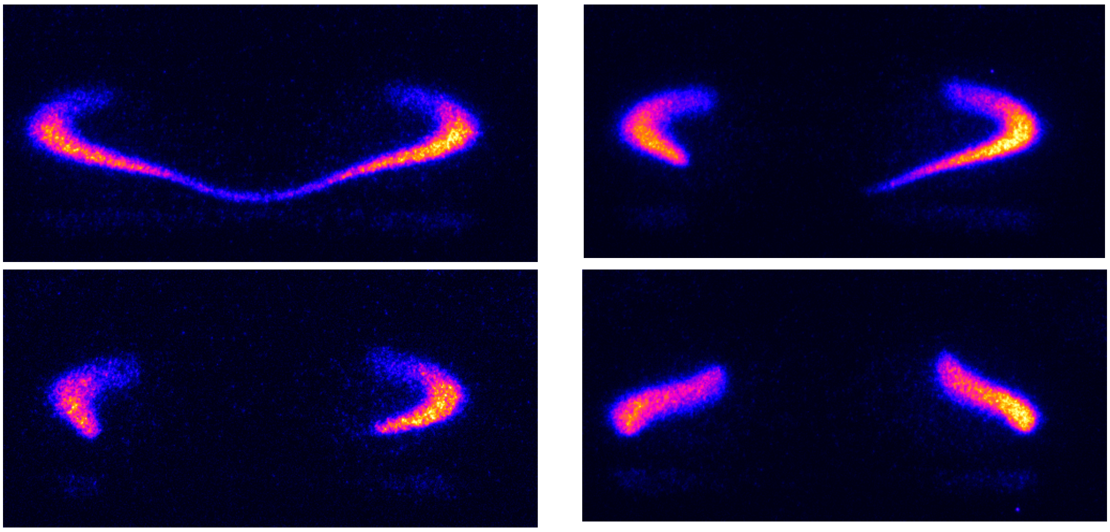

Figure 6: Single-shot 2-D OH LIF images from a methane-air flame, burner nozzles: 10mm dia. The initially burning diffusion flame (upper left) is perturbed by a sudden increase of the air velocity. The flame is quenched on the axis (upper right), the hole grows (lower left) and subsequently an edge flame is established as a new steady state (lower right).

## Nationale Zusammenarbeit

The experiments described here are planned in close collaboration with the *LAV* laboratory of *ETHZ*. Detailed numerical simulations are made there for some selected flame conditions, using boundary conditions which suit our experimental parameters.

## Internationale Zusammenarbeit

This project is included in the subtask 3.4M "Measurement of species concentration and temperature in combustion" of the IEA.

## Bewertung 2005 und Ausblick 2006

Using 1-D spontaneous Raman measurements, information on the structure of counter flow flames was gathered: Now two-dimensional temperature and major species concentration fields are available for a wide range of flame parameters (complementing the previously measured data base of

OH distributions and flow velocity fields). The spatially resolved information was applied to correct 2-D OH LIF measurements for collisional quenching. This constitutes a very important step towards the quantification of OH measurements - from LIF image to concentration fields.

Furthermore, the more detailed information available now also permits a more thorough comparison with numerical simulations performed at ETHZ, and an improved understanding resulted of the necessities required for accurate predictions.

The recording of transient flame structures met with unexpected problems. While the transition can be induced reliably, the timing of the laser measurement is not feasible with sufficient accuracy, due to a temporal jitter in the flame transition whose origin is not yet known. A different approach will be attempted with a high speed camera

## Referenzen

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