

Instantaneous and quantitative two-dimensional measurements of multiple scalars by Rayleigh/depolarized Rayleigh scattering in turbulent nonpremixed flames

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Abstract

We demonstrate the simultaneous, planar measurement of multiple scalars in a set of turbulent nonpremixed flames by polarized/depolarized Rayleigh-scattering. It uses correlations between signals and state variables (temperature, species concentrations). These correlations are taken from detailed multi-scalar measurements and from detailed numerical simulations. We show that many scalars can be approximated well as functions of the Rayleigh-signals. Using these functions, we can measure several scalars that otherwise would hardly be accessible to measurements, in a planar configuration, by one laser and two cameras. We reason why this method is likely to work well for a wide range of combustion systems and for other state variables, additionally to the ones shown here.

Introduction

Combustion involves a high number of scalar quantities that are required for its description, modelling and understanding. Laser-based experimental investigation techniques, *per se* can only deliver a small amount of these scalars (6-12 in typical Raman point or line measurements, only a few in planar measurements).

However, it is well established that the number of really independent quantities in a combustion process is actually quite small; this means that the process can be approximately described in terms of a few scalars: there is, at least approximately, an underlying reduced low-dimensional state space in which all quantities evolve (see e.g., [1, 2]). While this fact is readily used in numerical methods like CSP [3], ILDM [4], flame prolongation of ILDM [5] and FGM [6], it has, to our knowledge, not been systematically exploited so far in measurements.

The significance of the reduced state spaces for measurements is that the state variable correlations induce similar correlations between state variables and measurement signals. These correlations can be exploited in order to derive the values of state variables from measured signals. In this way, simultaneous measurements of multiple scalars with a small set of simultaneous measurements can become possible.

For this procedure to work, in principle the following requirements must be fulfilled: The reduced state space of the investigated system has to be known. Also, a mapping from state vectors to the measured signals is needed. This mapping, combined with the reduced state space, allows to establish correlations between signals and state variables. Finally, experiments must be conducted in order to measure signals, and these signals must be transformed into the corresponding values of state variables using the state signal correlations. As in every quantitative experiment, an error analysis is re-

quired to track the propagation of errors from the “raw” signals to the final results.

In this paper, we give a demonstration of a realization of these steps, by applying them to laser-based measurements in turbulent flames. As a means of establishing the reduced state space in these flames, we consider the TNF-Workshop nonpremixed piloted methane/air jet flames, which have readily been well characterized by extensive multi-scalar point measurements by Barlow and Frank [7] (measurement data are available for temperature, and mass fractions of CO₂, O₂, CO, N₂, CH₄, H₂O, H₂, OH, and NO [9]). These data are combined with detailed numerical simulations to ascertain the reduced state space that is occupied by the TNF jet flames.

The actual measurement signals used by us are two-dimensional images of the polarized and depolarized component of Rayleigh-scattered light, obtained after illumination with a high-power laser-sheet in the TNF D flame. Hereafter, we refer to these measurement signals as the “two Rayleigh-signals”. Since the relationship between the two Rayleigh-Signals and the state variables (temperature, pressure, mass fractions) is known quite accurately, we can calculate the two Rayleigh-signals from the point-measurement data. From the combined state variable and signal data, we are able to identify many sharp correlations between temperature, species concentrations and/or mass fractions and the two Rayleigh-signals. In other words, we can establish correlations between signals and many state variables.

By applying these correlations to measured images of the two Rayleigh-signals, we can simultaneously determine values of several scalars in a planar two-dimensional domain with a single laser shot and two cameras. This procedure is demonstrated here for temperature, concentrations of molecular oxygen, nitrogen and

methane, and also for mass fractions of water and carbon dioxide.

We also discuss the use of numerical simulation data to infer functional relationships between the signals and state variables. It is shown that, within the limits of experimental accuracy and for the above mentioned scalars, unstationary flamelets deliver the same functional dependence between signals and state variables as the measured data. Since this holds for the TNF flames C-F, covering a wide range of turbulence, this observation indicates that the method can be applicable also in flames where no detailed *a-priori* characterisation by multiscalar measurements are available.

Specific Objectives

We show that a systematic use of reduced state spaces, i.e., the use of correlations between measurement signals and state variables like temperature and species concentrations, can be a large aid in laser-based measurements in turbulent flames. We demonstrate that and how many quantities like temperature and species concentrations can be measured simultaneously, that are hardly accessible to other planar, quantitative measurements. We show that with two signals (namely the two Rayleigh-signals) we can measure temperature and several major species with good accuracy. This corresponds to a two-dimensional state space. Even more quantities may become accurately measurable if more dimensions (and therefore, more measurement signals) are regarded. This is the subject of future work.

Methodology

As outlined in the introduction, we first need to find the reduced state space of the investigated flames. To accomplish this, we consider a set of *a-priori* known state vectors that accurately represents the system under investigation. Of course, if a sufficient amount of accurately measured data is available, (“sufficient” with respect to the number of data points and the number of simultaneously measured quantities), they are a good choice for such a set. Among the experimentally and numerically best characterized combustion systems are the TNF-workshop nonpremixed piloted methane/air flames. Barlow and Frank provide a set of about 230000 data points, each containing the values of temperature T and mass fractions w_i for $i=CO_2, O_2, CO, N_2, CH_4, H_2O, H_2, OH, NO$, obtained from point measurements, with a spatial resolution of about 0.75mm [8, 9]. Measured data were considered from flames with different Reynolds numbers (namely the TNF-flames C-F, ranging from $Re \sim 13400$ to $Re \sim 44800$), from different heights above the burner exit, and different distances from the burner axis. More details of the measurements are given in the cited Barlow and Frank paper [8].

In addition to the measured point data, we sampled a set of approx. 30000 data points from unstationary, one-dimensional flamelet data. A counterflow configuration was considered. The length of the computational domain was 15mm, and Dirichlet-type boundary conditions were chosen for temperature, mass fractions and

velocities. To mimick the strongly unstationary flow-field in turbulent combustion, the tangential pressure gradient J (used for characterizing the strain rate) was varied with time during the calculations, according to a sinusoidal temporal profile, $J=J_0(1+A\sin(2\pi\omega t))$, where A ranged from 0 (no modulation) and 0.95 (large modulation), J_0 ranged from -10^4 N/m⁴ to -10^6 N/m⁴ and the frequency ω ranged from $100s^{-1}$ and $10000s^{-1}$. Note that the tangential pressure gradient is closely related to the velocity gradient (strain rate), [10]. The flamelet data included cases of complete extinction and of partial extinction with re-ignition. Flames were considered as being adiabatic, with a spatially and temporally uniform pressure ($p=1$ bar), and with a unity Lewis number assumption. The $Le=1$ assumption for our approach is justified *a posteriori*, see below. The unstationary flamelet data were used to provide a set of state vectors that could be compared to the measured data set with respect to correlations between state variables and measurement signals, as outlined below.

For each of the two data sets, Rayleigh and depolarized Rayleigh signals, I_r and I_d , resp., were calculated according to

$$I_r = K_r p M_m (R_{gas} T) \sum_i \sigma_i w_i / M_i$$

$$I_d = Tr(T) K_d p M_m (R_{gas} T) \sum_i \rho_i \sigma_i w_i / M_i$$

where p is the pressure, T the absolute temperature, M_m the molar mass of the gas mixture, $R_{gas}=8.314$ J/(mol·K) (universal gas constant), σ_i , ρ_i , w_i , M_i the Rayleigh scattering cross section, depolarization ratio, mass fraction and molar mass of species i , respectively. Summation is over all species. σ_i and ρ_i have been measured for the species $CH_4, O_2, H_2O, CO_2, CO, N_2, H_2$ and Ar; for the remaining species, the quantity $\sigma_i w_i$ was set to zero, i.e., the contribution of minor species to the Rayleigh-signals was neglected. The factors K_r and K_d were chosen such that I_r and I_d were equal to 1 for dry air at room temperature; this removed purely experimental parameters from the signals. $Tr(T)$ is a factor that accounts for the temperature dependent transmission of the depolarized signal through the detection line [11].

After calculation of the signals (I_r, I_d) as functions of the state vectors, we ascertain if points in (I_r, I_d, ψ)-space are correlated, where the symbol ψ represents any state variable (e.g. $\psi=T$, and or $\psi=w_i$). If so, we try to find an explicit underlying function $\psi=f(I_r, I_d)$ so that ψ can be determined later from measured values of the Rayleigh-signals (I_r, I_d).

This procedure is done for both the experimental and the unstationary flamelet data, i.e., we obtain a function $\psi=f_{exp}(I_r, I_d)$ from experiments and a (possibly different) function $\psi=f_{sim}(I_r, I_d)$ from simulations. By comparing the values of a particular state variable ψ that are obtained from f_{exp} and f_{sim} , we can judge how well the simulated and experimental correlations match.

The task of expressing the correlations can be expressed as explicit functions in a practically useful way is non-trivial. The correlations are given, both by the

experimental data and by the unstationary flamelets, as a set of scattered data points in (I_r, I_d, ψ) -space. In this “raw” form, the functions are not appropriate for use in practical evaluations. For instance, in measured images of I_r and I_d , there are typically around 50000 pairs of (I_r, I_d) given, (one pair at each pixel), for which the functions have to be evaluated to determine the corresponding ψ -field. The evaluation therefore should be fast and robust.

One way of accomplishing this is by mapping the scattered values (I_r, I_d, ψ) onto a regular, well-structured grid (e.g., a tensor product grid) in I_r, I_d space, so that fast interpolation routines can be used afterwards to determine ψ for a given value (I_r, I_d) . In this case, the gridded values (I_r, I_d, ψ) have to be stored. Typically, this involves 50×50 points per signal and state variable.

Another way is to find approximate analytical forms for the functions, e.g., by approximating ψ as a multivariate polynomial in I_r and I_d , or, more generally, a linear combination of arbitrary functions. Nonlinear parametric fits (i.e. fits where the fitting parameters enter nonlinearly into the function) might, in principle, be used as well.

We use approximation functions of the form $\psi = f(I_r, I_d) = A + BI_r + CI_d + D/I_r + E/I_d + F/(I_r + I_d) + \dots$, with parameters A, B, C, \dots that are determined by a linear least squares method. The “...” indicates the possible use of higher powers in I_r and I_d , as appropriate. These functions are chosen because they can describe a large range of different shapes, are linear in the fit-coefficients A, B, C, \dots , vary smoothly with I_r, I_d , are numerically well behaved for $I_r, I_d > 0$, and also fit well to the experimental and flamelet data in (I_r, I_d, ψ) space. The use of functions that are linear in the fit-coefficients is favorable since the fitting problem can then be reduced to solving an overdetermined linear equation system; for this task, robust and fast numerical algorithms exist. Moreover, these algorithms can guarantee to find the *global* best fit. If nonlinear terms of A, B, C, \dots would appear, it would be hard to find efficient, robust algorithms that can guarantee to find a global best fit.

Results and Discussion

Figure 1 shows as an example number densities of molecular oxygen ($[O_2]$) as a function of I_r and I_d . The underlying data are the experimental data set of Barlow and Frank and the unstationary flamelet data, as indicated in the caption. It is seen from this scatter plot that the point set $(I_r, I_d, [O_2])$ is “thin”, e.g., it does not fill the whole space, but forms approximately a surface.

The existence of low-dimensional surfaces in state-space is a consequence of the existence of low-dimensional surfaces (manifolds) in state space, and is therefore not surprising. What is remarkable is that the surface is to a very good approximation two-dimensional (it could be a hyper-surface in more dimensions), even though we consider a wide range of conditions in strongly turbulent (real turbulence for the measurement data, “mimicked” turbulence for the unstationary flamelet data) nonpremixed flames. Similar smooth two-

dimensional surfaces are found for temperature, number densities and/or mass or mole fractions of other major species (N_2, CH_4, CO_2, H_2O). Most other species (minor species) do not display such a smooth, sharp two-dimensional surface in (I_r, I_d, ψ) -space; they seem to require more than two dimensions in order to be accurately described in terms of a reduced state space. We restrict ourselves to two dimensions (corresponding to the two Rayleigh-signals) in this work.

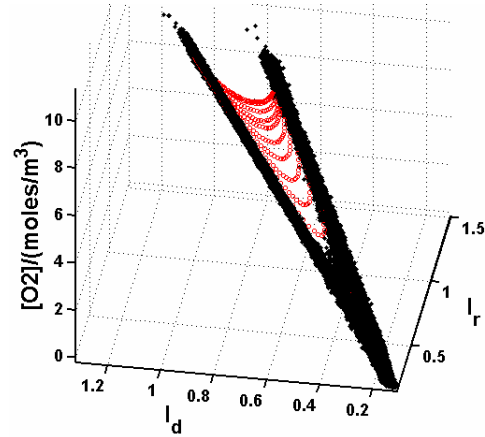


Figure 1 Points in $(I_r, I_d, [O_2])$ space from Barlow’s and Frank’s Raman/LIF experiments (9), dark full symbols) and from unstationary flamelets (open brighter symbols).

Comparing the experimental and the flamelet data, it is observed that both define the same underlying surface in $(I_r, I_d, [O_2])$ space. This is possibly not clearly visible in Figure 1 (three dimensional data are often hard to visualize on two-dimensional paper), but it can be confirmed quantitatively by reconstructing experimental $[O_2]$ values from the calculated signals $(I_r, I_d)_{exp.}$, using the *flamelet*-based surface $[O_2] = f_{fla}(I_r, I_d)$. We will show this further below, and similarly for T, N_2, CH_4, CO_2 and H_2O . First, we concern ourselves with determining the “thickness” of the surfaces.

In Figure 1, the flamelet data set appears to be slightly “thinner” than the experimental data set. We can get a more precise notion about “thinness” if we investigate the self-consistency of the data sets with respect to a two-dimensional surface. That means, we fit a smooth surface $f(I_r, I_d)$ through the data points $(I_r, I_d, [O_2])$, and then determine the deviation of all the values $[O_2]$ from the fitted surface $f(I_r, I_d)$. To visualize this deviation, we plot $[O_2]$ vs. the “reconstructed” value $[O_2]_{(rec.)} \equiv f(I_r, I_d)$. This is shown in Figure 2 for experimental and for flamelet data. The experimental data are more scattered, that is, they can less accurately be described by a smooth two-dimensional surface in state space. However, the relative standard deviation of the reconstructed $[O_2]$ values is not significantly larger (around 2% for room temperature, 10% for temperatures above 2000 K), than the relative standard deviation due to finite measurement precision. So, within the limits of available experimental precision, the data points are on a two-dimensional surface.

We could use the surfaces $f_{\text{exp}}(I_r, I_d)$ based on detailed point Raman/LIF measurements directly for the evaluation of the two Rayleigh-signals. However, it is more useful to have means of predicting the correlations for a given type of combustion systems from theory and numerical simulations, without the aid of detailed experiments. We therefore check if we can *reconstruct* experimentally determined values of certain scalars from the Rayleigh-signals, using correlations $f_{\text{fla}}(I_r, I_d)$ based on numerical simulations (unstationary flamelets). If this reconstruction is possible with a good accuracy, we can use the unstationary flamelet-based data instead of the experimental data for the determination of species. We check this by the following procedure:

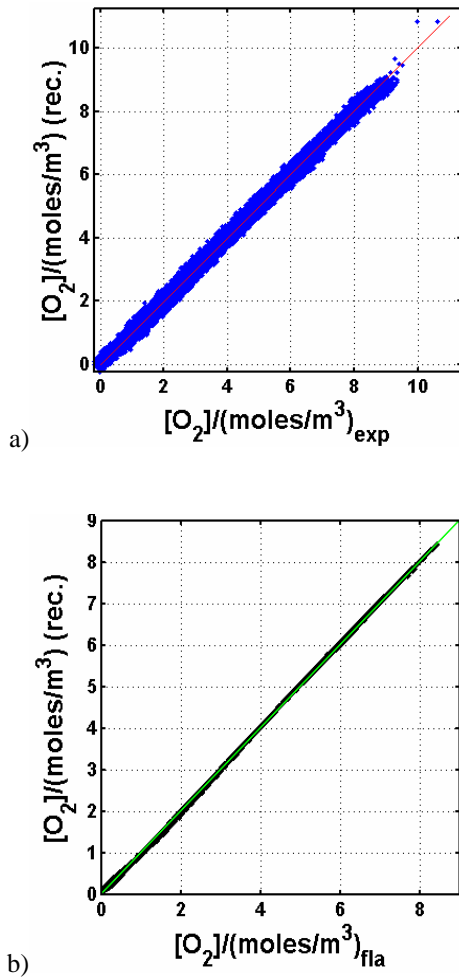


Figure 2 a) Reconstruction of experimental $[\text{O}_2]$ data by a smooth surface fitted through the $(I_r, I_d, [\text{O}_2])$ -data points. b) same for unstationary flamelet data.

We use the experimental set of state variables from Barlow's and Frank's Raman/LIF-measurements to calculate the corresponding Rayleigh-signals $(I_r, I_d)_{\text{exp}}$. It is then checked if/which state variables can be reconstructed in terms of the flamelet-based correlation $\psi_i=f_{\text{fla}}(I_r, I_d)$. Due to finite experimental precision, (and possibly also due to the fact that the real accessed region in (I_r, I_d, ψ_i) -space is maybe not exactly, but only

approximately a two-dimensional surface), there corresponds a set of reconstructed values $\psi_{i(\text{rec.})}^*$ to a certain experimental value of a state variable ψ_i^* . The important questions for measurements are (a) how well the mean of the reconstructed $\psi_{i(\text{rec.})}^*$ matches the "real" values ψ_i^* , and (b) how large the standard deviation from the mean of the $\psi_{i(\text{rec.})}^*$ is. (a) informs how well the flamelet-based and experiment-based correlations match and how well they are represented by the used fit-function. (b) informs about how well the data are approximated by a two-dimensional surface (compare the discussion above). The result for $\psi_i=[\text{O}_2]$ is shown in Figure 3. The x-axis represents the "real" experimental values, the y-axis represents the mean and \pm one standard deviation of all reconstructed values $\psi_{i(\text{rec.})}$. Note that the deviation between the mean of reconstructed values (dark full line) and the real values (red full line) is too small to be visible in most ranges, a slight deviation appears only around the region $[\text{O}_2]=6\dots 7 \text{ moles}/\text{m}^3$. Also, the standard deviation of the reconstructed values is tiny (relative standard deviation $< 3\%$ for $[\text{O}_2]=4 \text{ moles}/\text{m}^3$).

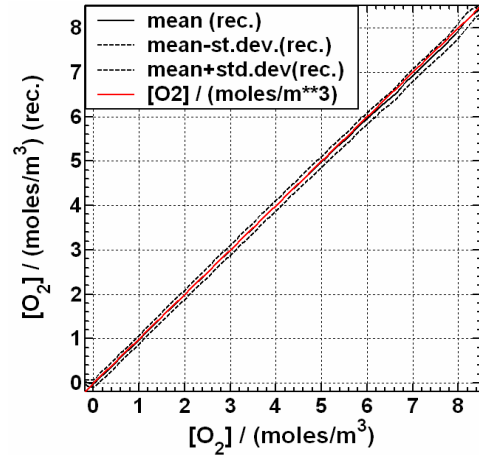


Figure 3 Mean (full line) and \pm one standard deviation (dashed lines) of $[\text{O}_2]$ values that reconstruct experimental data [9], using the correlation function from unstationary flamelets.

For T , $[\text{N}_2]$ and $[\text{CH}_4]$, the reconstruction of the experimental values by $\psi_i=f_{\text{fla}}(I_r, I_d)$ using the flamelet-based correlations works equally well, with standard deviations similar to the one for $[\text{O}_2]$. Mass fractions of water and CO_2 can also be reconstructed in the same way, although with somewhat larger scatter. The result for $w(\text{CO}_2)$ is shown as an example in Figure 4.

An important question is how errors in (I_r, I_d) propagate into the scalars. Since we could use only slightly nonlinear functions for the fits, the relative error propagation is benign, small relative errors in the signals result in nearly the same small relative errors in the reconstructed state variables. Generating a randomly distributed set of 10000 signals around the mean $I_r=0.16$ and $I_d=0.134$, (this corresponds to $T=1950 \text{ K}$, with a near stoichiometric mixture fraction), and with a relative standard deviation of 5% for I_r and I_d , we get the follow-

ing relative standard deviations of the reconstructed scalars: 6% for T , 5% for $[O_2]$, 5% for $[CH_4]$, 5% for $[N_2]$, and 6% for $w(CO_2)$ and $w(H_2O)$.

In practical applications, the *total* relative uncertainty in the value of the determined variable is the sum of the relative uncertainty due to the “unsharpness” of the underlying correlation (as seen in Figure 3 and Figure 4), and of the relative errors that are due to signal noise, as discussed above.

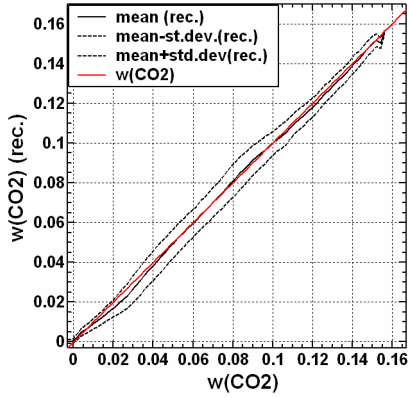


Figure 4 Reconstruction of experimental CO_2 mass fractions by flamelet-based correlations. Mean and standard deviation of reconstructed values are shown as in Figure 3.

Note that the correlations cover also cases where the physical disturbance of chemical reactions is strong, e.g., cases of full or partial extinction with re-ignition. Our method is therefore not limited to situations where turbulence-chemistry interactions are “weak”, but covers a wide range of Reynolds numbers.

Once the signal-state correlations have been established and cast into a numerically evaluable form, we can easily transform measured two-dimensional images of (I_r, I_d) into maps of state variables.

In Figure 5, maps of I_r, I_d and the corresponding reconstructed maps of $T, [O_2], [CH_4], w(CO_2)$ and $w(H_2O)$ are given. Flamelet-based surfaces were used for the reconstruction.

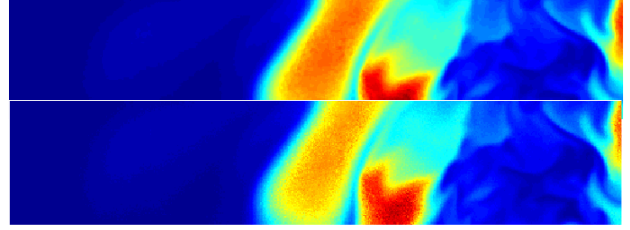
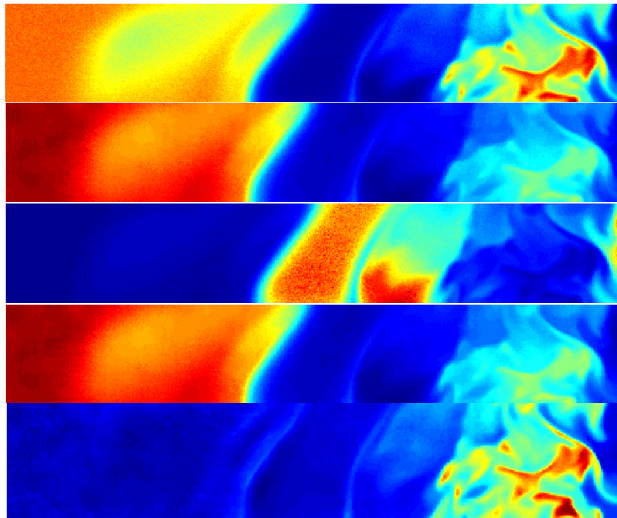


Figure 5 Maps (color coded, from blue to red) of various scalars. From top to bottom: Measured Rayleigh signals I_r and I_d , reconstructed values of T (color scale blue-red from 285-2100K), $[O_2]$ (0.2-8.7 mol/m³), $[CH_4]$ (0-10.5 mol/m³), $w(CO_2)$ (0-0.11), $w(H_2O)$ (0-0.13)

Note that $[O_2]$ is almost a linear function of I_d alone, therefore the maps for $[O_2]$ and I_d look very similar.

This procedure can be viewed as a special case of a more general approach for finding measurement strategies, based on reduced state spaces:

We search for measurement signals S that are known functions of the state variables (T, p, w_i) : $S = f(T, p, w_i)$. In our example, there was $S = (I_r, I_d)$, with $I_r = f_1(T, p, w_i) = K_1 p M_m / (R_{gas} T) \sum w_i / M_i \sigma_i$ and $I_d = f_2(T, p, w_i) = Tr(T) K_2 p M_m / (R_{gas} T) \sum w_i / M_i \sigma_i \rho_i$

Then, we try to assess correlations of the form $\psi = h(S)$ between state variables ψ ($\psi = T, p, w_i$) and the signals S by numerical simulations or, if possible, by experiments. In our example, we used numerical simulations and experimental data to find that the dimension was 2 for $\psi = (T, [CH_4], [O_2], [N_2], w(CO_2), w(H_2O))$.

Next, we bring these correlations into a practically evaluable form (e.g., by finding an appropriate analytical form or by creating a lookup-table). In our case, this was done by finding simple approximating functions to the $h(S)$.

Finally, we measure the signals S (e.g., in a planar configuration) and use $h(S)$ to convert them into state variables, using $\psi = h(S)$.

Conclusions

Even in strongly turbulent combustion, there exist sharp correlations between certain state variables and the intensities of Rayleigh and depolarized Rayleigh scattering.

As shown by a comparison of detailed experimental data in the TNF turbulent nonpremixed methane/air jet flames (by Barlow and Frank), and detailed unstationary flame simulations, the shape of many of these correlations can accurately be assessed by numerical simulations. Based on the correlations, a quantitative, planar measurement of multiple scalars, using one laser-sheet and two cameras (for recording the polarized and depolarized Rayleigh-signals) is possible with good accuracy (even excellent accuracy for the determination of $T, [O_2], [N_2], [CH_4]$). The studied cases cover a wide range of combustion scenarios with respect to turbulence/chemistry interaction; therefore, the results should apply to all flames with the same boundary conditions.

The fact that the reconstruction of experimental point-measurement data by unstationary flamelet-derived correlations works well for a set of state vari-

ables indicates that the $Le=1$ assumption is justified in our application, i.e., differential diffusion of heat or species in the studied flames does in these flames not affect the signal-state correlations.

This work is an example for the more general approach of using state variable/signal correlations for measurements in combustion. It has to be noted that this procedure is, at least in principle, almost guaranteed to be successful: It is well established by theory (e.g. QSSA, CSP, ILDM) that (at least approximate) low-dimensional correlations between the state variables exist. This is a well-understood consequence of the strongly different time scales in which the underlying chemical and physical processes occur. Since measurement signals can generally be written as functions of the state variables, the state variable correlations *have* to show up in a similar way as (low-dimensional) correlations between state variables and the signals. The question for the practical application is, what dimension (i.e., number of independent measurements) is required to get a reasonably sharp state-signal correlation, so that the equation $\psi=h(S)$ holds and can be brought into a practically useful form to determine ψ . The difficulty is mainly one of practical implementation.

We have the situation that likely a large number of measurement strategies, all based on reduced state spaces in a fashion similar to the one demonstrated here, exists; more work has to be done in order to find out which of these and how they can be best exploited for measurements in a practically useful way.

This paper is a first demonstration of this procedure. The fact that two measurement signals (from one laser-sheet) sufficed for a determination of at least 6 scalars in a nonpremixed, turbulent flame, indicates that this strategy is indeed promising, and that practically useful implementations are feasible.

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