The Role of Differential Diffusion during Early Flame Kernel Development under Engine Conditions - Part I: Analysis of the Heat-Release-Rate Response

Tobias Falkenstein\textsuperscript{a}, Aleksandra Rezchikova\textsuperscript{a}, Raymond Langer\textsuperscript{a}, Mathis Bode\textsuperscript{a}, Seongwon Kang\textsuperscript{b}, Heinz Pitsch\textsuperscript{a,∗}

\textsuperscript{a}Institute for Combustion Technology, RWTH Aachen University, 52056 Aachen, Germany

\textsuperscript{b}Department of Mechanical Engineering, Sogang University, Seoul 121-742, Republic of Korea

Abstract

Although experimental evidence for the correlation between early flame kernel development and cycle-to-cycle variations (CCV) in spark ignition (SI) engines was provided long ago, there is still a lack of fundamental understanding of early flame/turbulence interactions, and accurate models for full engine simulations do not exist. Since the flame kernel is initiated with small size, i.e. with large positive curvature, differential diffusion is expected to severely alter early flame growth in non-unity-Lewis-number (Le \neq 1) mixtures as typically used in engines. In this work, a DNS database of developing iso-octane/air flame kernels and planar flames has been established with flame conditions representative for stoichiometric engine part-load operation. Differential diffusion effects on the global heat release rate are analyzed by relating the present findings to equivalent flames computed in the Le = 1

∗Corresponding author

Email address: office@itv.rwth-aachen.de (Heinz Pitsch)
limit. It is shown that in the early kernel development phase, the normal propagation velocity is significantly reduced with detrimental consequences on the global burning rate of the flame kernel. Besides this impact on the overall mass burning rate, the initial production of flame surface area by the normal propagation term in the flame area balance equation is noticeably reduced. By using the optimal estimator concept, it is shown that strong fluctuations in local heat release rate inherent to Le $\neq 1$ flames in the thin reaction zones regime are mainly contained in the parameters local equivalence ratio, enthalpy, and H-radical mass fraction. Differential diffusion couples the evolution of these parameters to the unsteady flame geometry and structure, which is analyzed in Part II of the present study (Falkenstein et al., Combust. Flame, 2019).

Keywords: Flame Kernel, Differential Diffusion, Flame Stretch, DNS, Premixed Flame, Spark-Ignition Engine

1. Introduction

Combustion stability is a prerequisite for more efficient spark ignition (SI) engine operation [2, 3] and reduced engine-out emissions [4, 5]. The occurrence of cycle-to-cycle variations (CCV) is mainly attributed to the very early combustion phase [6], which may take up to 30% of the entire combustion duration to oxidize less than 2% of the in-cylinder fuel mass [7]. By using advanced laser-optical diagnostics, the time until the young laminar flame kernel transitions to a turbulent flame (at approx. 1% burned mass) was shown to correlate with CCV [8]. Further, it was found that stretch effects in Le $\neq 1$ mixtures affect the duration until 10% of fuel are consumed, which
correlates with CCV as well. Although differential diffusion effects have been systematically investigated in spherical expanding turbulent flames by several experimental research groups [9–12], limited accessibility of the smallest time- and length scales demands for complementary numerical analyses.

While flame kernels developing in turbulent flow fields have been computed in various DNS studies [13–21], differential diffusion effects have been considered mainly in recent years [22–30]. A non-exhaustive list of DNS parameters and flame conditions that have been investigated to date is provided in Tab. S-1 of the supplementary material.

In the early DNS study by Echekki et al. [22], a Lewis number variation was performed for the very first time in a flame kernel configuration. It was found that flame growth is significantly accelerated in a mixture with $Le = 0.53$ compared to $Le = 1$ conditions. More detailed insights were enabled only many years later by increasing computational power and availability of high-performance computing resources. Dunstan and Jenkins [23] investigated the effect of hydrogen enrichment on the development of lean methane/air flame kernels, in analogy to a previous planar-flame study by Hawkes and Chen [31]. The behavior of pure methane/air flames was shown to be similar in both configurations. By contrast, hydrogen enrichment yielded a stronger enhancement of the flame kernel turbulent burning velocity due to the stronger impact of reduced thermal-diffusive stability in presence of higher global stretch rate. Dinesh et al. [27] performed DNS of lean H$_2$/CO/air (syngas) flame kernels at two Reynolds numbers with $Le = 0.385$ (realistic) and $Le = 1$. At elevated turbulence levels, the significant increase in flame area caused by thermal-diffusive effects observed at
the lower Reynolds number was found to diminish. Differential diffusion was shown to increase the local burning velocity, hence accelerating flame kernel growth, even at the higher Reynolds (and moderate Karlovitz) number. Alqallaf et al. \[30\] systematically varied the Lewis number (\(Le = 0.8, 1.0, 1.2\)) in DNS of flame kernel development computed with single-step chemistry to analyze the effect on flame curvature dynamics. Flame propagation was shown to smoothen the flame surface for \(Le \geq 1\), while production of both positive and negative curvatures is promoted in mixtures with \(Le < 1\).

Extinction of an \(H_2/air\) ignition kernel, i.e. before the critical radius for self-sustaining flame propagation is reached, was studied by Uranakara et al. \[28\]. A particle-based analysis was utilized to understand the effect of turbulence-enhanced heat loss leading to extinction. Saito et al. \[29\] performed two-dimensional DNS to study the effect of small-scale turbulence on the ignition of methane/air and \(n\)-heptane/air flame kernels. After normalization with the laminar flame and ignition delay times, similar sensitivities to strain were observed for both mixtures.

Regarding DNS studies on developed turbulent flames, those which were performed under engine-relevant conditions or those which considered larger hydrocarbon fuel species are particularly relevant for the present work. Savard et al. \[32\] analyzed effects of pressure on turbulent, slightly lean (\(\phi = 0.9\)) iso-octane/air flames at different Karlovitz numbers. Laminar chemical reaction pathways primarily contributing to heat release were significantly changed by ambient pressure. It was found that at low pressures of 1 bar, strong variations in heat release were caused by higher sensitivity to curvature due to differential diffusion as compared to 20 bar. Increased Karlovitz numbers
lead to reduced fluctuations in heat release due to enhanced turbulent species transport inside the flame structure.

Aspden et al. [33] computed lean dodecane/air flames for a range of Karlovitz numbers ($Ka = 1$-36) in the thin reaction zones regime. Small-scale turbulent mixing was found to cause significant deviations in reaction rates from laminar flames computed with $Le = 1$ and $Le \neq 1$, which is in contrast to methane/air flames [34]. In the low-temperature region of the dodecane/air flame structure, reaction rates of fuel and intermediate species were shown to be significantly reduced at elevated Karlovitz numbers, which was attributed to turbulent perturbations of the fuel breakdown. This effect was identified as a distinct feature of heavy-hydrocarbon flames due to the spatial separation of fuel consumption and heat release.

Although significant progress has been made towards more realistic DNS of premixed flames using detailed chemistry, investigations on the behavior of actual road transportation fuels under engine-relevant turbulence and thermodynamic conditions are still very scarce. While few studies on canonical planar flame configurations exist in literature, the role of molecular transport, as well as its interaction with small-scale turbulent mixing and chemistry inside the flame structure of flame kernels has not been studied in a realistic setting. Experimental evidence on the relevance of early flame kernel development for CCV in SI engines and the lack of accurate models suggest to address this gap in literature. In the first part of the present study, the heat-release-rate response to differential diffusion effects is analyzed based on the integrated chemical source term, which has high practical relevance, and based on the local chemical source term, which is used to identify the
governing parameters in Le ≠ 1 flames in a quantitative fashion. In the second part, the coupling between the local mixture state, which determines the local heat release rate, and the flame geometry and structure is established.

The present manuscript is organized as follows. In Sect. 2 the overall analytical approach pursued to explore differential diffusion effects in the present engine-relevant datasets is summarized. A brief overview of the DNS database is provided in Sect. 3. The global and local heat release rate are analyzed in Sect. 4 and suitable parameters for a reduced representation of differential diffusion phenomena inside the flame structure are proposed.

2. Analytical Approach

To assess the overall combustion process in technical combustion devices (e.g. in SI engines), global parameters are typically of interest, such as the volume-integrated heat release rate or reaction progress variable source term:

$$\overline{\dot{\omega}_c}|_{\Omega} = \frac{1}{V_\Omega} \int_{\Omega} \dot{\omega}_c \, dV.$$  \hspace{1cm} (1)

Here, $c$ is a synonym for a reaction progress variable, e.g. a (normalized) temperature or a quantity representative for the major product species. In spark ignition engines, a fast burning rate is desirable mainly for three reasons. First, thermal efficiency is improved when approaching the limit of constant-volume combustion. Second, reduced residence times of the end gas (e.g. near hot surfaces) achieved by faster arrival of the flame front may reduce knock tendency. Third, an acceleration of the early flame kernel growth shortens the transition time to a fully developed turbulent flame \[7,8\]. This makes the young flame less prone to stochastic kernel/flow interactions, which may
significantly reduce CCV. However, differential diffusion effects may severely slow down the early flame kernel development for mixtures of common transportation fuels and air with \( \text{Le} > 1 \).

To give a first impression of the impact of differential diffusion under engine conditions, the integrated reaction source term has been evaluated in a stoichiometric iso-octane/air flame kernel DNS dataset, and an equivalent dataset computed in the \( \text{Le} = 1 \) limit. In Fig. 1 it is shown that \( \text{Le} = 1 \) overall leads to a substantially higher burning rate than in case of the realistic engine fuel, despite a 16\% lower \textit{unstretched} laminar burning velocity. To estimate the influence on CCV in actual engines, the laminar-to-turbulent transition time \( \tau_{\text{lam-turb}} \) proposed by Schiffmann et al. [8] has been evaluated for both flames. It turns out that the presence of differential diffusion leads to a 31\% increase in \( \tau_{\text{lam-turb}} \), which may increase CCV in terms of the coefficient of variance of the mean effective pressure \( \text{COV}_{\text{IMEP}} \) from 1.8 to 4.9\% (cf. Fig. 7 in [8]).

To gain a general understanding of the complex processes inside the flame
structure of turbulent flames and enable the derivation of accurate models, it is desirable to identify a suitable reduced representation of the multi-parameter system and to establish a connection to canonical laminar reference flames. In the present work, this is pursued by following two analysis pathways according to Fig. 2.

Starting point is $\bar{\omega}_c|_\Omega$ as quantity of interest (QoI) for engine combustion. In a first step, the flames will be considered from a macroscopic perspective that allows to intuitively demonstrate the impact of differential diffusion on early flame kernel development within the frame of the flame surface density (FSD) concept [35]:

$$\bar{\omega}_c|_\Omega = (\rho s_{l,0}) \cdot \bar{I}_0 \cdot \Sigma_{c,\Omega}, \quad (2)$$

where $\rho$ is the fluid density, $\Sigma_{c,\Omega} = \bar{A}_c/V_\Omega$ [36] denotes the global flame surface density and the stretch factor $\bar{I}_0$ quantifies deviations of the displacement speed $s_d$ from the laminar burning velocity $s_{l,0}$ due to ignition, differential diffusion, and turbulent micro-mixing. To parametrize $\bar{\omega}_c|_\Omega$ entirely in terms of flame structure and geometry, $\bar{I}_0$ can be decomposed into contributions by normal propagation, which is affected by the flame structure, and by tangential diffusion, which represents the flame geometry [37]:

$$\bar{I}_0 = \frac{\langle \rho s_d \rangle_{s,\Omega}}{\rho s_{l,0}} = \frac{\langle \rho (s_{rn} - D_{th} \kappa) \rangle_{s,\Omega}}{\rho s_{l,0}}$$

$$=: \bar{I}_{0,rn} + \bar{I}_{0,\kappa}, \quad (3)$$

where $\langle \cdot \rangle_s$ denotes generalized scalar-isosurface averaging [36], $s_{rn}$ is the displacement speed due to reaction and normal diffusion, and $D_{th}$ is the thermal diffusivity. In this work, the mean curvature $\kappa$ of iso-surfaces belonging to
any scalar field $\vartheta$ is computed from the normal vector $n_i$ pointing into the
direction of negative scalar gradient, i.e. flame kernels have positive global
mean curvature:

$$
\kappa^{(\vartheta)} = \frac{\partial n_i^{(\vartheta)}}{\partial x_i},
$$

(4)

$$
n_i^{(\vartheta)} = -\frac{1}{|\nabla \vartheta|} \frac{\partial \vartheta}{\partial x_i}.
$$

(5)

For brevity, the sub/superscript indicating the scalar field will be omitted in
most parts below.

The macroscopic perspective based on the reduced flame representation
in terms of a propagating front (cf. 1 in Fig. 2) enables a separate consider-
ation of differential diffusion effects on $I_0, r_n$ (flame structure and the chemical
source term) on the one hand, and on $I_0, \kappa$ as well as $\Sigma_c, \Omega$ (flame geometry)
on the other hand. In this way, the actual displacement speed of early flame
kernels can be related to a laminar unstretched flame and the transition
from a quasi-laminar kernel to a fully developed turbulent flame front may
be quantified by the evolution of total flame area $\bar{A}$, which is directly related
to the curvature distribution of the flame [44]. While this perspective already
captures the external effect of hydrodynamic strain, a differentiated consid-
eration of energy supply by spark ignition, as well as effects of turbulent
micro-mixing on the coupled multi-species system inside the flame structure
require an additional analysis pathway.

Starting point for the micro-scale analysis is the local chemical source
term $\dot{\omega}_c$ (cf. 2 in Fig. 2). In contrast to the displacement speed, the source
term only depends on the local state vector $[Y_k, T]$, which shall be expressed
by a reduced parameter set in order to characterize differential diffusion ef-
Figure 2: Reduced representation of turbulence/flame interactions on the integrated progress variable source term \( \overline{\omega_c} \) in presence of differential diffusion (\( \text{Le}_k \neq 1 \)). Two analysis pathways are suggested:  

1. A macroscopic perspective based on the FSD concept (cf. r.h.s. of \( (\overline{\omega_c}) \)-Eq. in the blue box) considers a propagating flame front with deviations in flame displacement speed from an unstretched laminar flame, similar to laboratory experiments or classical asymptotic theories [45, 46].  
2. A micro-scale perspective on the local source term \( \dot{\omega}_c \), which is determined by the local mixture state, characterized by the local equivalence ratio \( \phi \), enthalpy \( h \), and radical mass fraction \( Y_{\text{radical}} \). In case of \( \text{Le}_k \neq 1 \), these parameters change according to the local flame structure (\( |\nabla c| \)) and geometry (\( \kappa \)), which leads to a response in \( \dot{\omega}_c \) to external perturbations of the flame, e.g. by turbulence. Note that turbulent fluctuations in the scalar fields ahead of the flame have been omitted.
fects. As will be shown in Sect. 4.2, the combination of the local equivalence ratio $\phi$, local enthalpy $h$, and the mass fraction of a radical species is well-suited to capture variations in $\dot{\omega}_c$. The evolution of each parameter is determined by the respective transport equation, but only the (approximate) differential diffusion term $D_{Le}$ of the enthalpy equation is given in Fig. 2 for brevity. Although the present $D_{Le}$-formulation may not be valid for high-Karlovitz-number conditions, it highlights the importance of local flame structure ($|\nabla c|$) or local geometry ($\kappa$) for the occurrence of deviations from laminar flame behavior in case of $Le_k \neq 1$. For more details, refer to Part II of the present study [1]. From the $\kappa$- and $|\nabla c|$-Eqs. given in Fig. 2 it is immediately obvious that local flame geometry and structure are coupled, i.e. externally invoked changes in $\kappa$ or $|\nabla c|$ (e.g. by hydrodynamic strain), may alter the respective other parameter. Overall, the presence of differential diffusion effects ($Le_k \neq 1$) leads to a response in heat release rate to externally introduced perturbations in local flame geometry and structure, which may in turn change the $\kappa$ and $|\nabla c|$ balance. Note that in the limit of $Le_k = 1$, the local mixture state will be much less affected and heat release response can be expected to be small.

While run-to-run variations between different flame kernel realizations computed in the $Le_k = 1$ limit were investigated in our previous studies in terms of the $(\overline{\omega}_c)$- and $(\overline{A})$-Eq. [38] as well as the $(\kappa)$-Eq. [44], the objective of the present work is to isolate the effect of differential diffusion on $\overline{\omega}_c$ during early flame kernel development. In Part I of the present study, the global heat release rate is first analyzed from the macroscopic perspective (r.h.s. of $(\overline{\omega}_c)$-Eq. in Fig. 2) to compare the behavior of a flame kernel computed
with a realistic engine fuel to previous results computed in the $\text{Le}_k = 1$ limit. Then, the heat release rate is considered from a micro-scale point of view to identify the parameters that characterize the coupling between $\dot{\omega}_c$ and turbulence/flame interactions in presence of differential diffusion. In Part II of the present study [1], the effect of the local flame structure and geometry on the mixture state $(\phi, h, Y_H)$ and source term $\dot{\omega}_c$ is quantitatively investigated by relating the behavior of flame kernels to corresponding planar turbulent flames and laminar reference solutions.

3. DNS Database

In order to fundamentally characterize early flame kernel development under SI engine conditions, four flame kernel DNS realizations and one planar flame with $\text{Le} > 1$ were added to an existing database of two reference flames computed in the $\text{Le} = 1$ limit (cf. Sect. 3.1). Additional comparative analyses are enabled by laminar unstretched and stretched flamelet solutions, which will be summarized in Sect. 3.2.

3.1. Three-Dimensional DNS

The DNS database has been carefully designed to be representative for SI engine part load conditions. Since the governing equations and numerical methods were extensively described in our previous study on the same flame configurations as in this work, but with all Lewis numbers artificially set to unity [38, 44], only a brief summary is provided in the following. To enable systematic investigations on differential diffusion effects, four flame kernel realizations and one planar reference flame with realistic (but constant) Lewis numbers have been added to the existing $\text{Le} = 1$ database. Hence, four
Table 1: Flame conditions in the DNS.

| Property            | Value                      |
|---------------------|----------------------------|
| Mixture             | Iso-Octane/Air             |
| $p^{(0)}$           | 6 bar                     |
| $T_u$               | 600 K                     |
| $\phi_u$           | 1.0 (homogeneous)         |
| $s_l^o$             | 0.73 m/s                  |
| $l_t$               | 69.1 $\mu$m               |
| Flow Field          | Decaying h.i.t.            |
| Combust. Regime     | Thin Rct. Zones           |
| Le_{eff}            | 2.0 | 1.0 |

flame configurations are available for this study. While the flame conditions listed in Tab. 1 are fully equivalent to our previous work [38, 44], some differences in the non-dimensional groups given in Tab. 2 emerge due to the smaller chemical time scale of the laminar unstretched flame with Le > 1. In Tab. 1, the mixture thermodynamic state is given by the pressure $p^{(0)}$ and temperature of the unburned gas $T_u$ with equivalence ratio $\phi_u$. Note that the turbulent flow field corresponds to decaying homogeneous isotropic turbulence, which explains the DNS parameter ranges given in Tab. 2. The turbulent integral length scale and eddy turnover time are denoted by $l_t$ and $\tau_t$, $\eta$ is the Kolmogorov length scale, while the laminar flame thickness and initial flame diameter are referred to as $l_f$ and $D_0$, respectively. Due
Table 2: Engine [47, 48] and DNS characteristic numbers (Le > 1).

| Parameter | Engine | DNS (t\text{init} - t\text{end}) |
|-----------|--------|---------------------------------|
| Re\text{t} | 100 – 2390 | 385 – 222 |
| \frac{u_{rms}}{S_l} | 2 – 15 | 5.9 – 2.8 |
| Ka | 1 – 6 | 10.6 – 3.2 |
| Da | 1 – 100 | 1.9 – 4.6 |
| \frac{t_d}{\eta} | 100 – 200 | 87.2 – 57.2 |
| \frac{t_d}{\ell} | 20 – 147 | 10.9 – 13.0 |
| \frac{D_n}{\ell} | < 1.0 | 0.3 | ∞ |

to computational restrictions, the wavenumber range covered by the DNS is smaller than in an actual engine. Hence, the integral length scale in the simulations is two to three times smaller than in reality, while the Karlovitz number is closely matched to actual engine conditions. For an illustration of the DNS conditions in the regime diagram of turbulent combustion, refer to [38]. Although the Karlovitz number in that study was slightly higher than in the present work due to the Le = 1 simplification, it was shown that no thickening of the averaged flame structure occurs.

In order to quantitatively assess the impact of differential diffusion on the heat release rate, a detailed chemical reaction scheme is required [49]. As in our previous study, a modified kinetic model based on the skeletal iso-octane mechanism by Pitsch and Peters [50] was used. Validation results for the
reaction scheme calibrated to the present DNS conditions are provided as supplementary material.

A brief summary of the DNS setup is given in Tab. One important difference between the flame kernel datasets and the planar reference flames is the initialization method. Flame kernels were ignited by a source term in the temperature equation, which results in an early growth phase comparable to engine experiments reported in literature (cf. [44]). By contrast, a laminar unstretched flame was imposed into the turbulent flow field as initial condition for the planar-flame DNS to avoid strong dilatation due to the larger burned volume.

To simplify the analysis and make the dataset more accessible for modelling, a new reaction progress variable $\zeta$ has been defined by the solution of the transport equation [38]

$$
\frac{\partial}{\partial t} (\rho \zeta) + \frac{\partial}{\partial x_j} (\rho u_j \zeta) = \frac{\partial}{\partial x_j} \left( \rho D_{th} \frac{\partial \zeta}{\partial x_j} \right) + \dot{\omega}_\zeta. 
$$

The chemical source term was chosen as the sum of source terms of main product species:

$$
\dot{\omega}_\zeta = \dot{\omega}_{H_2} + \dot{\omega}_{H_2O} + \dot{\omega}_{CO} + \dot{\omega}_{CO_2}. 
$$

Solving Eq. (6) in the DNS simplifies the analysis of the overall normal and tangential diffusion effects, as compared to a progress variable definition that results in a balance equation with more complex diffusive transport terms. The decomposition of the first term in the r.h.s of Eq. (6) according to Echekki and Chen [37] will be employed in Sect. 4.1 to analyze the decomposed stretch factor (cf. Eq. 3).
| Property                      | Value                                      |
|-------------------------------|--------------------------------------------|
| Grid Size                    | $960^3$                                    |
| Domain Size                  | $15 \cdot l_t$                             |
| Navier-Stokes Eq.            | Low-Mach-Approx. [51]                      |
| Transport Model              | Curt.-H. [52], const.-Le                   |
| Soret Effect                 | yes                                        |
| Chem. Mechanism              | 26 Spec., based on [50]                    |
| Flame Kernel Init.           | Ignition Heat Source                       |
| Sim. Time of Flame-Kernel    | $1x \ (t_{\text{sim.}} = 3.4 \cdot \tau_t)$, |
| Kernel Realizations          | $3x \ (t_{\text{sim.}} = 1.0 \cdot \tau_t)$ |

Table 3: Computational setup of the DNS.
3.2. Laminar Flame Calculations

To relate local conditions inside the turbulent flame structures to laminar flamelet solutions, several one-dimensional flames were computed with the FlameMaster code [53]. Steady, unstretched laminar flames (sub/superscript ‘lam’) were solved based on the algorithm by Smooke et al. [54]. To generate stretched reference flames, the premixed back-to-back counterflow configuration (sub/superscript ‘cff’) has been selected in analogy to previous modeling studies [55, 56]. The numerical solutions were obtained based on the similarity coordinate formulation assuming a potential flow in the far field [57]. For reaction mechanism validation and assessment of early flame kernel growth, laminar spherical flames (sub/superscript ‘sph’) have been computed in Lagrangian coordinates [58] using DASSL [59].

A detailed analysis of the laminar unstretched flame structure in iso-octane/air flames in terms of a very similar kinetic scheme as used in the present study was provided by Pitsch and Peters [50] and is not repeated here. For a discussion of differential diffusion effects on flame structure and extinction in the premixed counterflow configuration, refer to the modeling study by van Oijen and de Goey [60] or the review paper by Lipatnikov and Chomiak [61].

4. Results

As described in Fig. 2, the analysis of heat-release-rate response has been conducted according to two logical pathways. In Sect. 4.1, differential diffusion effects on the global heat release rate will be discussed from the macroscopic perspective by considering two equivalent flame kernel datasets com-
puted with Le > 1 and Le = 1, respectively. For the micro-scale analysis, the ability of several reduced parameter sets to capture differential-diffusion-induced fluctuations in the local heat release rate of planar flames will be quantified in Sect. 4.2. The main results will be used to explain the Lewis number effects on the global heat release rate of flame kernels observed in Fig. 1.

4.1. Global Heat Release Rate Evolution

To better understand the strong impact of differential diffusion on $\dot{\omega}_c|_\Omega$ that was shown in Fig. 1, the stretch factor $I_0$ and the flame surface density $\Sigma_{c,\Omega}$ will be considered according to the r.h.s. of the $(\dot{\omega}_c)$-Eq. in Fig. 2. In this way, differential diffusion effects on the local burning velocity can be distinguished from effects on flame area evolution. Note that the results in Fig. 1 were plotted as function of time, which is relevant for the engine application. However, this representation implies that differences in the integrated source term accumulate due to growing differences in flame kernel size. In order to rigorously analyze the role of differential diffusion during flame development, it is desirable to enable comparisons of data extracted from flames with similar geometry and size. Hence, results will be shown as function of kernel radius hereafter. The radius is here defined as the median of the distance distribution, measured between all flame points and the geometric center of the flame.

A comparison of the stretch factors evaluated for both the Le > 1 and the Le = 1 turbulent flame kernel is provided in Fig. 3(a). Initially, $I_0$ is dominated by the effect of spark ignition, which affects the early flame kernels until a radius of approximately one integral length scale is reached. This
behavior is similar to actual engines, as discussed in our previous study [44]. At the kernel size of one integral length scale, differential diffusion effects reduce $I_0$ by 33%. This is a stronger reduction than observed for two laminar spherical expanding flames, which were ignited in the same way as the DNS and are plotted in Fig. 3(a) for reference (superscript ‘sph’). As shown in Part II of the present study [1], turbulence detrimentally alters the flame structure by hydrodynamic strain, in addition to the unfavorable effect of large positive curvature on the heat release rate of $Le > 1$ flame kernels. It should be noted that $I_0$ quantifies the deviation of the actual flame displacement speed from the respective laminar burning velocity, which is in fact lower in case of $Le = 1$ ($s^n_{1,Le=1}/s^n_{1,Le>1} = 0.84$). At the kernel size of one integral length scale, the net displacement speed of the $Le > 1$ flame is 21% lower than in the $Le = 1$ limit.

Decomposing the stretch factor into a component representative of flame normal propagation and a curvature effect (cf. Eq. (3)) provides additional insights. Obviously, the normal-propagation stretch factor $I_{0,n}$ is significantly reduced in the $Le \neq 1$ flame after ignition effects have decayed. According to the $(s_n)$-Eq. in Fig. 2 this can be due to changes in the chemical source term $\dot{\omega}_c$ and the scalar gradient magnitude $|\nabla c|$, which is related to the local flame thickness. From the definition of $I_{0,\kappa}$ (cf. Eq. (3)), it can be expected that differential diffusion only has a minor effect on this term, since the correlation between curvature and the diffusion coefficient is rather weak and mean curvature is not altered by changes in Lewis number. This is confirmed by the results shown in Fig. 3(a). Note that $I_{0,\kappa}$ is a measure for diffusive transport of $\zeta$ in iso-surface-tangential directions, while curvature effects on
the chemical source term are contained in $I_{0,\text{rn}}$.

The domain-integrated generalized flame surface density $\Sigma_{\zeta,\Omega}$ is plotted in Fig. 3(b) as function of kernel radius. Obviously, the realistic engine fuel leads to a suppression of net flame area production due to thermal-diffusive effects, which is in agreement with planar flame results reported in literature [62–64]. When the flame kernels have reached a radius of one integral length scale, which corresponds to the time when ignition effects have mostly decayed [44], the flame surface density of the $Le > 1$ flame is reduced by 15% compared to the $Le = 1$ case. Note that both turbulent flames exhibit significantly larger surface areas than a perfect sphere with the same radius, as indicated in the figure. From the preceding discussion it can be concluded that the reduction in integral heat release rate attributed to differential diffusion effects is initially dominated by the burning velocity, while noticeable differences in flame area exist as well.

In order to further investigate how differential diffusion reduces the overall flame surface area (or $\Sigma_{\zeta,\Omega}$) and consequently the integral heat release rate
of the Le > 1 flame kernel (cf. Fig. 3(b)), the total flame area rate-of-change is considered [38]:

\[
\frac{1}{A_{\zeta, \Omega}} \frac{D_{\zeta} (\bar{A}_{\zeta, \Omega})}{D_{\zeta} (t)} = \frac{\int_{\Omega} (s_{\text{m}} \kappa - D_{\text{th}} \kappa^2 + a_t) |\nabla \zeta| \, dV}{\int_{\Omega} |\nabla \zeta| \, dV}.
\]

(8)

Results extracted from the Le > 1 and the Le = 1 datasets are compared as function of kernel size in Fig. 4. First, we focus on the evolution of the l.h.s. of Eq. (8) to assess differences in net area rate-of-change. As shown in Fig. 4(a), flame area production is overall positive due to the presence of mean flame curvature, but generally higher in the Le = 1 case. At \((R_{50} = 1.0 \cdot l_t)\), when ignition effects have mostly decayed, differential diffusion leads to a reduction in flame area production rate by 25\%. From the strong reduction in flame normal-propagation velocity observed in Fig. 3(a), it can be expected that the first term in the r.h.s. of Eq. (8), i.e. the change in flame area due to normal propagation of a curved surface, plays a major role in causing this difference. Hence, this term is plotted separately from the tangential strain and scalar dissipation terms in Fig. 4(b). In this way, it becomes obvious that the normal-propagation term is responsible for almost all differences in early flame area growth in the range \((R_{50} < 1.0 \cdot l_t)\). In particular, the sum of the remaining two terms is initially very similar for both Lewis number cases, while differential diffusion reduces the area production through normal propagation by up to 50\%. During this early phase, the effect of differential diffusion on flame kernel area growth may be similar to laminar flames (cf. Fig. 3(a)). As the flame kernels grow in size, differences in the curvature distributions develop (cf. Part II [1]), which cause differences in heat release rate that feed back into the tangential strain term in Eq. (8).

The preceding discussion has shown that differential diffusion has a strong
impact on the burning rate during flame kernel development under engine-
relevant conditions especially because of its influence on flame normal propa-
gation. Both flame area and flame displacement speed are significantly lower
in the flame computed with a realistic transportation fuel, i.e. with $Le > 1$.
Compared to an unstretched laminar flame, the net displacement speed is
reduced by up to 40 % due to differential and tangential diffusion. As this
minimum in displacement speed occurs at very small flame radii, the overall
flame kernel behavior may still be very sensitive to stochastic interactions
with the turbulent flow field [44]. Hence, the discussed burning-rate evolu-
tion is expected to be particularly critical for the occurrence of CCV in SI
engines.

4.2. Local Heat Release Rate Variations

To further investigate the strong Lewis-number dependence of the mean
normal-propagation stretch factor $I_{0,\text{rn}}$ (cf. Sect. 4.1), the parameter interac-
tions inside the flame structure that govern the local heat release rate will
be analyzed in the following. Since the physical and chemical phenomena
inside an unsteady flame structure are already quite complex, the following analysis is simplified by limiting ourselves to the fully developed, statistically planar-turbulent-flame datasets. It will be shown that in addition to a reduction in mean heat release rate, tremendous fluctuations in heat release rate exist throughout the flame structure of the $\text{Le} > 1$ flame. To parametrize the heat release rate in $\text{Le} \neq 1$ flames, the combination of local equivalence ratio, enthalpy, and radical availability will be quantitatively compared to other parameter choices and confirmed as suitable parameter set, in agreement with individual findings from literature that were used to design Fig. 2.

To get a first impression of the impact of differential diffusion on the spatial heat release distribution, the temperature iso-surfaces corresponding to maximum heat release rate in the laminar unstretched flame (‘$T_{\text{maxHR}}$’) have been extracted from both planar-turbulent-flame datasets at $t = 2.8 \tau_t$ and are colored by heat release rate in Fig. 5. In the $\text{Le} = 1$ dataset, the heat release rate distribution is almost uniform. Conversely, the more engine-relevant flame with $\text{Le} > 1$ features similarly high source term magnitudes only in regions of negative curvature, while large parts of the plotted iso-surface exhibit significantly reduced heat release rates. This observation is consistent with the correlation between the fuel consumption rate and curvature that was previously shown for an n-heptane/air flame in the broken reaction zones regime [65] and the correlation between the consumption speed and curvature in $\text{Le} \neq 1$ flames located in the corrugated flamelet regime [62]. Correlations between the local heat release rate and flame structure/geometry are discussed in Part II of the present study [1]. For a visualization of differential diffusion effects during flame kernel development, refer...
Figure 5: Planar Flame: $T_{\text{maxHR}}$ iso-surface colored by local heat release rate for the $Le = 1$ dataset (a) and the engine-relevant $Le > 1$ flame (b) at $t = 2.8 \tau_l$.

to Figs. S-3 and S-4 of the supplementary material.

To enable a more quantitative understanding of differential diffusion effects on the heat release rate distribution across the flame structure, joint-PDFs of heat release rate and temperature are shown in Fig. 6. Additionally, conditional mean results are plotted for comparison with laminar reference data. In the $Le = 1$ flame, the heat release rate of an unstretched premixed flame is almost perfectly recovered (cf. Fig. 6(a)). In the $Le > 1$ case, a different behavior is expected due to the reduction in stretch factor that was observed in the corresponding flame kernel dataset (cf. Fig. 3(a)). As shown in Fig. 6(b), the maximum conditional mean heat release rate of the turbulent planar flame is reduced by 29% with respect to the laminar unstretched value. For reference, data extracted from a back-to-back counterflow flame (c.f.f.) solution has been added to the figure. The
strained laminar flame solution has been selected based on the condition $|\nabla T|_{\text{maxHR,cff}} = \langle |\nabla T| \mid (T = T_{\text{maxHR}}, \kappa = 0) \rangle$ and will be analyzed in more detail in Part II [1]. The difference between the turbulent and the laminar profiles on the high-temperature side of the peak can be reduced by conditioning the data on zero curvature, which is not shown for clarity.

Besides the reduction in mean heat release rate observed in the planar, $Le \neq 1$ flame as compared to the corresponding $Le = 1$ dataset, visual inspection of Fig. 6 immediately suggests a strong impact of differential diffusion on variations in local heat release rate. While the data points in the $Le = 1$ case shown in Fig. 6(a) are distributed in a rather narrow band around the conditional mean profile, tremendous scatter from almost extinguished flamelets to levels well above the unstretched reference flame is present in the $Le > 1$ dataset. Note that the occurrence of flame elements with particularly low heat release rates was previously identified as a distinct feature of the thin reaction zones regime [66].

As a next step, we seek to identify the parameters that govern the local
heat-release-rate response to differential diffusion effects, i.e. the observed high conditional variance and reduced conditional mean. The findings will be used to relate the detrimental reduction in global heat release rate due to $\text{Le} > 1$ (cf. Fig. 1) to the mixture conditions inside the flame structure. To this end, we make use of the optimal estimator analysis technique \[67, 68\].

The primary objective of the following parameter tests is not to derive or check a specific model, but to quantify the ability of a selected input parameter set to predict local heat release rate throughout the flame structure, irrespective of a functional model expression. In this regard, the optimal estimator concept is a powerful tool which is based on two main ideas. First, the conditional mean of a dataset is considered as an ideal reference model, i.e. the optimal estimator. Second, the expected error of a model based on the selected input parameters $\Pi$ cannot be smaller than the standard deviation from the respective optimal estimator, which is cast into an error measure called the irreducible error:

$$\epsilon^2(\Pi) = \| \dot{\hat{\omega}}_T - \| \dot{\hat{\omega}}_T \| \Pi \| \| \Pi \| .$$ (9)

To reduce fitting errors which may pollute the optimal estimator results, artificial neural networks have been employed as suggested by Berger et al. \[69\]. Since we are specifically interested in the role of differential diffusion, all irreducible errors have been normalized by $\epsilon^2_{\text{Le}=1}(T)$, i.e. the error value computed for the $\text{Le} = 1$ flame when using temperature as the only model parameter. Hence, a normalized error value of unity would imply that the scatter around the conditional mean yields the same error sum as computed from the data points plotted in Fig. 6(a). The expected irreducible errors associated to heat release rate predictions from different input param-

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eter sets in the Le > 1 flame are listed in Tab. 4. When only temperature is used to parametrize heat release rate (cf. Fig. 6(b)), the irreducible error is 5.5 times larger than in the Le = 1 flame.

The empirical input parameter selection closely follows the schematic shown in Fig. 2 in a backward-analysis fashion, starting from the local heat release rate \( \dot{\omega}_T \) as quantity of interest. From a chemical kinetic point of view it might be interesting to begin the backward analysis with the identification of pathways that differ between the Le \( \neq 1 \) and the Le = 1 flames. Similar analyses have been published for different fuels [70, 71], including systematic variations of the Lewis number [72], but are not pursued here. Still, finding suitable marker species which contain most of the observed heat release rate variations may serve as a starting point for future work and may suggest control variables for flamelet modeling. Due to the direct influence of individual species mass fractions on chemical source terms, these are here referred to as first-level parameters (reaction/diffusion-controlled). Among common marker species used for modeling of hydrocarbon flames [55], molecular hydrogen was found to yield an irreducible error \( \epsilon^2 (T, Y_{H_2}) \) comparable to the reference error \( \epsilon^2_{Le=1} (T) \).

As a next step, heat release rate has been parametrized by the stoichiometry and energy content of the local mixture. The chosen second-level parameters (diffusion-controlled) for a reduced representation of the mixture state are local equivalence ratio [73, 74] and enthalpy [75], which are not changed by chemical source terms. Recall that the unburned mixture is homogeneous, i.e. variations in both parameters can only occur due to differential and thermodiffusion. Although enthalpy and equivalence ratio are
not independent quantities [60], $e^2(T, \phi, h)$ is clearly beneficial compared to using only one of both parameters. Note that the irreducible error $e^2(T, h)$ is larger than $e^2(T, \phi)$, which seems in contradiction to the early work by Ashurst et al. [75], who identified enthalpy as a suitable parameter to characterize flame dynamics in $Le \neq 1$ mixtures. This discrepancy to the present results is likely a feature of the multi-species system as opposed to single-step-chemistry DNS. The irreducible error can be further reduced by supplementing the second-level parameter set by one radical mass fraction [10, 76, 77]. In particular, $e^2(T, \phi, h, Y_{H_2})$ yields the smallest normalized error value of all $Le > 1$ tests. In fact, this parameter set gives the same errors in both the $Le > 1$ and $Le = 1$ flames, which is in agreement with the combination of individual findings reported in literature (cf. Fig. 2). Although molecular hydrogen was identified as the most suitable first-level-parameter, the combination $(T, \phi, h, Y_{H_2})$ is less effective than considering radical availability, which might be due to a correlation between $Y_{H_2}$ and $\phi$.

As enthalpy and equivalence ratio variations are linked to flame curvature and scalar gradient magnitudes through $D_{Le}$ (cf. Fig. 2), $\kappa$ and $|\nabla T|$ form the third-level parameters (kinematic) for a reduced description of the flame geometry and structure. For simplicity, only the temperature field has been considered to evaluate both parameters, which is a reasonable approximation in the present datasets (cf. Part II [1]). However, the approximate expression for $D_{Le}$ given in Fig. 2 is certainly inexact and will result in an increased irreducible error. While accounting for curvature in $e^2(T, \kappa)$ leads to some reduction in the expected irreducible error, the dominant third-level parameter seems to be $|\nabla T|$. Although both quantities are not independent [78],
the parameter combination yields a lower error \( \epsilon^2(T, \kappa, |\nabla T|) \), which is still more than 50\% higher than the reference error \( \epsilon^2_{Le=1}(T) \). It should be noted that such deterioration in performance of an ideal model is expected as the relation between the quantity of interest (heat release rate) and input parameters becomes weaker. This increased variance compared to the \( Le = 1 \) flame might be due to local changes in species mass fractions and gradients induced by turbulent micro-mixing (i.e. the temperature field may be less representative) or finite response time of the second-level (diffusion-controlled) parameters \( (\phi, h) \) to changes in curvature and scalar gradients. The latter effect is investigated by testing a fourth-level parameter set (kinematic/dynamic), which contains an additional quantity representative for the time rate-of-change of the third-level parameters. Here, the tangential strain rate has been selected as a measure for hydrodynamic changes to the flame structure (cf. (|\nabla T|)-Eq. in Fig. 2) and geometry. In fact, a correlation between local enthalpy and strain rate was already shown to exist in the earliest DNS with \( Le \neq 1 \) [75]. Since it is well-known that curvature and strain are correlated [62, 79], it is not surprising that the irreducible errors \( \epsilon^2(T, |\nabla T|, a_t) \) and \( \epsilon^2(T, |\nabla T|, \kappa) \) are identical. Still, accounting for both curvature and tangential strain in addition to \( |\nabla T| \) is even more effective. An almost identical irreducible error is achieved by replacing the tangential strain rate by the gradient rate-of-change (l.h.s. of (|\nabla T|)-Eq. in Fig. 2) to include the flame structure dynamics into \( \epsilon^2 \left( T, \kappa, |\nabla T|, \frac{1}{|\nabla T|} \frac{\partial T(\nabla T)}{\partial t} \right) \). The optimal estimator analysis has quantitatively shown that at a given progress variable (temperature), preferential diffusion effects on the heat release rate are well represented by the parameter set \( (\phi, h, Y_{H}) \). When the data is conditioned on
Table 4: Irreducible error intrinsic to heat release rate predictions from different input parameter sets (normalized by the temperature-based error value computed from the $L_e = 1$ flame).

| Parameter Set II | $\epsilon_{Le>1}^2(\Pi) / \epsilon_{Le=1}^2(T)$ |
|------------------|-----------------------------------------------|
| $T$              | 5.49                                          |
| $T, Y_{H_2}$     | 0.93                                          |
| $T, Y_{H}$       | 1.84                                          |
| $T, h$           | 1.88                                          |
| $T, h, Y_{H}$    | 1.07                                          |
| $T, h, Y_{H_2}$  | 0.77                                          |
| $T, \phi$        | 0.94                                          |
| $T, \phi, Y_{H}$ | 0.50                                          |
| $T, \phi, Y_{H_2}$ | 0.76                               |
| $T, \phi, h$     | 0.64                                          |
| $T, \phi, h, Y_{H}$ | 0.30                              |
| $T, \phi, h, Y_{H_2}$ | 0.55                               |
| (Le = 1 : $T, \phi, h, Y_{H}$) | (0.31)                             |
| $T, \kappa$      | 3.84                                          |
| $T, |\nabla T|$   | 1.78                                          |
| $T, |\nabla T|, \kappa$ | 1.52                                          |
| $T, a_t$         | 4.68                                          |
| $T, \kappa, a_t$ | 3.46                                          |
| $T, |\nabla T|, a_t$  | 1.52                                          |
| $T, \kappa, |\nabla T|, a_t$  | 1.35                                          |
| $T, \kappa, |\nabla T|, \frac{1}{|\nabla T|} \frac{\partial r(|\nabla T|)}{\partial r(t)}$ | 1.36 |
Figure 7: Planar Flame: Joint-PDFs of heat release rate and temperature for the Le = 1 dataset (a) and the engine-relevant Le > 1 flame (b) at $t = 2.8 \tau_l$. All data has been conditioned on \((\langle \phi | T \rangle, \langle h | T \rangle, \langle Y_H | T \rangle)\).

this reduced representation of the local mixture state, the remaining scatter around the conditional mean heat release rate is similar for both Le > 1 and Le = 1 as shown in Fig. 7. The coupling between \((\phi, h, Y_H)\) and the third-level (kinematic) parameters \((\kappa, |\nabla T|)\) as well as the role of external effects (e.g. turbulence, cf. Fig. 2), will be analyzed in Part II of the present study [1].

To finally connect the presented macroscopic and micro-scale analyses, the correlation between the local mixture state parameters $h$ and $\phi$ with $I_{0,\text{rn}}$ (cf. Fig. 3(a)) during flame development will be briefly discussed hereafter. Both $h$ and $\phi$ have been averaged over the reaction zones of the $\text{Le} \neq 1$ flames and are plotted in Fig. 8 as function of time. Note that the planar flame was initialized as a laminar unstretched flame, which features slightly rich mixture in the reaction zone. By contrast, the flame kernel evolves from a pocket of excess enthalpy. Consequently, the flame kernel enthalpy drops below the enthalpy level of the planar flame not before $t = 1.2 \tau_l$ as shown in Fig. 8(a), while the mixture composition in both flames approaches
Figure 8: Reaction-zone-averaged enthalpy (a) and local equivalence ratio (b) as function of time for \( \text{Le} \neq 1 \). Note that these are approximately constant in \( \text{Le} = 1 \) flames.

the minimum equivalence ratio level already at \( t = 0.6 \tau_t \) (cf. Fig. 8(b)). The combination of both effects influences the local burning velocity evolution (cf. Fig. 3(a)). Regarding differences between flame geometries, the flame kernel shows a minimum local equivalence ratio well below the stationary level of the planar flame, which is expected due to the positive mean curvature.

Since small flame kernels are particularly sensitive to external perturbations, the reduction in burning velocity (cf. Fig. 3(a)) caused by low enthalpy and equivalence ratio inside the reaction zone in \( \text{Le} > 1 \) mixtures is critical in terms of CCV in engines. However, systematic development of an appropriate spark ignition strategy may enable effective counter measures. From laminar investigations it is known that the minimum ignition energy (MIE) increases with increasing Lewis number \([80, 81]\), which is in line with the present turbulent flame results.
Conclusions

The intention of the present work is to clarify the role of differential diffusion during early flame kernel development in the thin reaction zones regime. Due to the expected relevance for the occurrence of cycle-to-cycle variations in SI engines, a DNS database has been carefully designed to be representative of practical engine conditions. Conclusive analyses were enabled by systematic variations of the global flame geometry and the mixture Lewis number of a realistic transportation fuel and air. A macroscopic analysis of the global burning rate as practical quantity of interest has shown the impact of differential diffusion during early flame kernel development. Further, the local heat release rate response has been used as starting point for a systematic micro-scale analysis on the complex parameter dependencies inside the turbulent flame structure. Specifically, the following conclusions can be drawn:

- The detrimental effect of differential diffusion on the net burning rate of flame kernels is mainly due to changes in the normal propagation velocity, while differences in flame area at the same flame radius are slightly less important, though still significant under the present conditions. This is in agreement with the findings of Dinesh et al. [27] in flames with Le < 1. However, the observation that the flame-area dependence on thermal-diffusive effects is weakened under high-Reynolds-number flow conditions [27] may not translate to the very early phase of flame kernel development, which is a topic suggested for future work.

- Early flame kernel area growth is significantly reduced in the Le > 1
flame, which has been attributed to a reduction in the normal-propagation term in the surface area evolution equation. Area production by propagation of the curved flame surface is proportional to the normal-propagation stretch factor \( I_{0,m} \), which has been shown to strongly suffer from low enthalpy and local equivalence ratio levels inside the reaction zone as a consequence of differential diffusion. Under the present turbulent conditions, differential diffusion thus acts on initial flame area growth in a very similar way as known from laminar spherical flames.

- Non-unity Lewis numbers were shown to result in strong variations in local heat release rate. In particular, very low heat release rates were locally observed. This is in agreement with results reported by Shim et al. [66] for H\(_2\)/air flames located in the thin reaction zones regime. However, such locally low heat release rates were neither observed under flame conditions in the corrugated flamelets regime by Shim et al., nor in the present \( Le = 1 \) datasets in the thin reaction zones regime. Hence, heat release fluctuations seem to be particularly pronounced in \( Le \neq 1 \) flames located in the thin reaction zones regime. Both of these conditions are typically found in SI engines.

- By using the optimal estimator concept, the fluctuations in heat release rate for a given temperature (or progress variable) have been quantitatively attributed to different parameter dependencies and related to the behavior of a \( Le = 1 \) flame. Specifically, a hierarchical grouping of parameter sets based on coupling-strength with the quantity of interest (heat release rate) has been proposed, i.e. first-level (reaction/diffusion-
controlled), second-level (diffusion-controlled), third-level (kinematic) and fourth-level (kinematic/dynamic) parameters. On each level, a dominant parameter can be identified (cf. Tab. 4), which may be useful for modeling purposes. For the present analysis, the objective was to find a reduced representation of the local mixture state, which determines the heat release rate. It turned out that conditioning on the parameter set \((T, \phi, h, Y_H)\) reduces the remaining scatter around the conditional mean heat release rate to identical levels in both \(Le > 1\) and \(Le = 1\) flames, which is in agreement with the proposed flame physics schematic based on findings from literature (cf. Fig. 2).

The coupled nature of the local mixture state with the flame geometry and structure due to differential diffusion is discussed in Part II of the present study [1].

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