A multi-scale asymptotic scaling and regime analysis of flamelet equations including tangential diffusion effects for laminar and turbulent flames

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\textbf{A B S T R A C T}

Recently, the relevance of tangential diffusion effects has been identified for laminar flames. These effects are not considered in the classical flamelet equations. In the present work, flamelet equations including these effects are derived, and their relevance is investigated by a multi-scale asymptotic scaling analysis. The analysis yields characteristic ratios dependent on the local curvature of the mixture fraction field, the scalar dissipation rate, and the flame thickness, which indicate whether tangential diffusion effects become important. By comparing relevant scales, a regime diagram is developed and three different flamelet regimes are identified. In regime I, the classical flamelet equations are valid. In regime II, differential diffusion of species and temperature in flame-tangential direction becomes relevant. In regime III, additional transport along mixture fraction isosurfaces exhibits an influence on the flame structure. In the latter case it is not sufficient to condition species mass fractions and temperature on the mixture fraction alone since these quantities are not represented by a one-dimensional structure.

The asymptotic scaling is verified against fully resolved numerical data of a laminar non-premixed methane–air flame and a turbulent lifted hydrogen jet flame. Budgets of different flamelet solutions of the laminar flame reveal that tangential diffusion effects are dominant over the classical flamelet terms near the flame centerline, while in regions away from it, standard flamelet terms are prevailing. For the turbulent flame, the results show that tangential diffusion effects are more localized and can exceed flame-normal transport, although, unsteadiness is more likely the key factor of the configuration.

While the classical flamelet model is applicable for certain flames, this study shows that tangential diffusion effects may require consideration for general flame configurations.

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\section{Introduction}

The utilization of flamelet models for the simulation of chemically reacting flows offers a substantial reduction of computational cost when using complex chemical-kinetics mechanisms. The main requirement for its application is, that chemical time scales are at least an order of magnitude smaller compared to the characteristic time scales of the flow field. This corresponds to the assumption that the Damköhler number of the rate-limiting chemical reaction is large, or – equivalently – that chemistry is fast. As a consequence, chemical reactions are confined to thin reaction–diffusion layers, and this matter is commonly represented by the thin flame-sheet assumption. Based on this assumption, it was shown by Peters\textsuperscript{[1]} that throughout the reaction zone one-dimensional structures, so-called flamelets, can be identified. Therefore, the governing equations for species mass fractions and temperature can be transformed into a one-dimensional form. Instead of solving the flamelet equations at simulation run-time, flamelet solutions can be precomputed. Hence, thermochemical data can be tabulated as a function of a reduced set of scalars, which results in significant speed-up of the simulation.

The classical one-dimensional flamelet formulation\textsuperscript{[2]} was derived for equal species and thermal diffusivities corresponding to unity Lewis-numbers ($\text{Le} = 1$). The Lewis number is defined as:

\begin{equation}
\text{Le} = \frac{\lambda}{\rho c_p D},
\end{equation}

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where \( \lambda \) represents the thermal diffusivity, \( \rho \) the density, \( c_p \) the heat capacity, and \( D \) the diffusivity. Considerable progress has been made in extending the combustion-physical representation of the flamelet model, thereby enabling its application to increasingly more complex flame configurations. In particular, Pitsch and Peters [3] extended the flamelet equations by including differential diffusion effects (Le \( \neq 1 \)). Furthermore, the laminar flamelet concept extends to turbulent combustion, for which the unity Lewis number assumption is often an adequate approximation since molecular transport is small compared to turbulent diffusivities [2].

Despite its successful application to various non-premixed flame configurations, limitations of the flamelet formulations were found in previous work [4–9]. Kortschik et al. [4] experimentally identified significant curvature effects studying autoignition in corrugated counterflow mixing fields. They were the first to introduce a curvature-based correction term into the flamelet equations trying to overcome limitations of the classical flamelet models. Other authors [5–9] found shortcomings of flamelet models studying a laminar non-premixed co-flow flame that was originally reported by Bennett et al. [10]. Claramunt et al. [5] compared different mathematical formulations of the flamelet approach to a detailed chemistry solution. Their results showed that classical steady flamelet solutions are able to predict main flame properties. However, they often poorly describe intermediate species and overestimate the temperature on the lean part of the flame. Furthermore, Claramunt et al. [5] concluded that it is important to include differential diffusion and possibly unsteady effects to capture relevant flame properties correctly. More recently, Verhoeven et al. [8] showed that incorporating non-unity Lewis number effects into the flamelet formulation improves the results significantly. From one-dimensional calculations they found that flamelet models are able to reproduce planar chemical flame structures including differential diffusion. Nevertheless, they pointed out that curvature-induced tangential diffusion effects near the axis of the flame lead to significant deviations, since these effects are not accounted for in the classical flamelet equations. Xuan and Blanquart [11] investigated an axisymmetric co-axial diffusion flame in an attempt to predict sooting tendencies by applying a classical one-dimensional flamelet model. They found that this model is inadequate in reproducing the correct species profiles along the flame centerline. They argued that this failure is due to the omission of multi-dimensional convection and diffusion and introduced modified flamelet equations. Xu et al. [12] analyzed the flamelet structure along the centerline of an axisymmetric reacting particle in a convective flow. They derived flamelet equations that contain additional terms to account for curvature effects for cases with unity and non-unity Lewis numbers. A budget analysis showed that curvature terms become relevant for both cases especially in the wake region of the particle. Furthermore, their results showed that the consideration of these additional terms provides substantially improved results, while classical flamelet models for Le = 1 and Le=1 fail to accurately predict species and temperature profiles. Xuan et al. [13] presented a similar set of equations as derived by Xu et al. [12] for conditions away from the axis. They showed that their one-dimensional flamelet model, including some, but not all of the tangential diffusion effects, improves the results. Another approach was introduced by Nguyen et al. [14] who discussed multi-dimensional flamelet-generated manifolds. In their model, temperature and species mass fractions are conditioned on the mixture fraction \( Z \) and the progress variable \( Y_i \). They show that multi-dimensional effects, corresponding to fluxes across iso-equivalence-ratio and iso-progress-of-reaction surfaces, can hardly be fully neglected in chemistry tabulation of partially premixed combustion.

The objective of this work is the derivation of flamelet equations including tangential diffusion effects through an asymptotic scaling analysis utilizing multiple small scales. From this analysis, a regime diagram is developed that provides a rigorous description of the relevance of additional terms in the flamelet equations. The applicability of this flamelet formation is then examined by considering two different flame configurations: a laminar non-premixed methane–air flame [10] and a turbulent lifted hydrogen jet flame [15].

The remainder of this paper has the following structure. In Section 2, we review the origin of tangential diffusion effects from a physical perspective. The asymptotic scaling and regime analysis and the derivation of the flamelet equations are presented in Section 3. Results from this analysis are given in Section 4, and the paper finishes with conclusions in Section 5.

### 2. A physical description of tangential diffusion effects

Tangential diffusion effects generally consist of two different types: (i) transport along mixture fraction isosurfaces, also denoted as multi-dimensional effect and (ii) differential diffusion in flame-tangential direction. In the following the origin of these effects is explained from a physical perspective.

By considering the species and mixture fraction equations for the special case of unity Lewis numbers, the only difference between the equations is the chemical source term

\[
\rho \frac{\partial Y_i}{\partial t} + \rho \vec{v} \cdot \nabla Y_i = \nabla \cdot (\rho D_Z \nabla Y_i) + \dot{m}_i, \tag{2}
\]

\[
\rho \frac{\partial Z}{\partial t} + \rho \vec{v} \cdot \nabla Z = \nabla \cdot (\rho D_Z \nabla Z). \tag{3}
\]

When assuming a two-stream chemically reacting flow, species at the oxidizer side will diffuse towards the fuel side and vice versa. Further, the species mass fractions and mixture fraction, are advected by the same flow field \( \vec{v} \). Hence, it can be concluded that outside the reaction zone, where all chemical source terms vanish, species mass fraction and mixture fraction fields tend to align.

In case that the characteristic chemical time scale is small compared to the convective and diffusive time scales, combustion takes place in an asymptotically thin layer. In the limit of infinitely fast chemistry the reaction zone thickness tends to zero. Since the mixture fraction and species mass fraction fields are assumed to align outside the reaction zone, they will consequently align everywhere if the reaction zone is infinitely thin. Therefore, \( Y_i \) can be conditioned on the mixture fraction \( Z \), which will lead to the classical flamelet equations when transforming Eq. (2) into mixture fraction space [1].

The scalar dissipation rate, defined as \( \chi = 2D_Z |\nabla Z|^2 \), will influence chemical reactions, eventually quenching the flame if increased above a critical value. Since the scalar dissipation rate represents the inverse of a characteristic diffusion time, this can be considered as a reaction–diffusion interaction within the reaction zone. Therefore, a large local scalar dissipation rate can be interpreted as a short residence time in the reaction zone. Considering

**Fig. 1.** Multi-dimensional effects arise from a changing scalar dissipation rate \( \chi \) while \( Z \) remains constant. This introduces gradients \( \partial Y_i /\partial s \) and \( \partial T /\partial s \), where \( s \) is the coordinate measured along the \( Z \) isoline.
two points on a curved Z isoline in the reaction zone (see Fig. 1), the scalar dissipation rate might change along this line, while Z remains constant. Assuming finite rate chemistry it follows that gradients of $Y_i$ and $T$ exist along the Z isoline, which cannot be resolved by the flamelet model through conditioning $Y_i$ and $T$ on Z alone. In the present work, the diffusive transport that occurs due to these gradients is denoted as a multi-dimensional effect.

Figure 2 shows geometrical conditions between isolines of $Z$ and $Y_i$ that illustrate when this effect becomes relevant. Multi-dimensional effects are not relevant for planar flames (see Fig. 2a) and for spherical symmetric flames (see Fig. 2b), due to the perfect alignment of $Z$ and $Y_i$ isolines. For an axisymmetric flame (see Fig. 2c), however, multi-dimensional effects might require consideration. This case has been studied by Xu et al. [12] who derived flamelet equations including multi-dimensional effects for the axial flamelet of an axisymmetric reacting coal particle. The flamelet equations for the species mass fractions assuming unity Lewis numbers write [12]:

$$\rho \frac{\partial Y_i}{\partial t} = \rho \frac{\partial Y_i}{\partial z} \left( \frac{D_{Y_i}}{2} \right)^2 \left( K_{Y_i} - \bar{K}_Y \right) \frac{\partial Y_i}{\partial z} + \bar{m}_i,$$

where the curvatures $K_{Y_i}$ and $\bar{K}_Y$ are defined as

$$K_{Y_i} = -\nabla \cdot \bar{n}_{z} = -\nabla \left( \frac{\nabla Z}{|\nabla Z|} \right) = -\frac{\nabla^2 Z}{|\nabla Z|} - \nabla \left( \bar{n}_{z} \cdot \nabla Z \right),$$

$$\bar{K}_Y = -\nabla \cdot \bar{n}_Y \frac{\partial Y_i}{\partial z}.$$

The second term on the right hand side (RHS) of Eq. (4) can be interpreted as transport along the $Z$ isoline. For the case that the curvatures $K_{Y_i}$ and $\bar{K}_Y$ are zero or that the scalar fields of $Y_i$ and $Z$ are perfectly aligned, the multi-dimensional effect vanishes. Therefore, it is concluded that this effect is dependent on the misalignment of both isolines. Further, it is noted that the equations do not hold when considering the most general case in Fig. 2d, as simplifications due to symmetry on the axis cannot be incorporated any longer and the gradients of $Y_i$ and $Z$ are misaligned.

Taking into account differential diffusion effects (non-unity Lewis numbers) introduces additional tangential diffusion terms [12]. Two of these four additional terms in the species flamelet equation are related to transport along mixture fraction isosurfaces and are therefore multi-dimensional effects. The remaining two terms depend on the mixture fraction curvature $K_{Z}$ and originate from differential diffusion along the flame-tangential direction. Somewhat similar equations have been derived by Xuan et al. [13] and were incorporated into a one-dimensional flamelet model. Although they neglected multi-dimensional effects, their model showed reasonable agreement with CFD results for a laminar co-flow flame. In the following we will show that the omission of multi-dimensional effects is insufficient for the laminar co-flow flame examined in this work.

It is the scope of the following analysis to derive a flamelet formulation that is applicable to general diffusion flame configurations. Therefore, differential diffusion will be considered in the derivation of the flamelet equations by employing a non-unity Lewis number approach that has successfully been applied by Xu et al. [12]. While the mixture fraction is described by Eq. (3) the governing equations for species and temperature read:

$$\rho \frac{\partial Y_i}{\partial t} + \rho \bar{v} \cdot \nabla Y_i = \nabla \cdot \left( \rho D_{Y_i} \nabla Y_i - Y_i \sum_{k=1}^{n} \rho D_{k} \nabla Y_k \right) + \bar{m}_i,$$

$$\rho \frac{\partial T}{\partial t} + \rho \bar{v} \cdot \nabla T = \frac{1}{c_p} \frac{\partial p}{\partial t} + \nabla \cdot \left( \frac{\bar{\lambda}_c}{c_p} \nabla T \right) + \frac{\bar{\lambda}_c}{c_p} \nabla \bar{c}_p \cdot \nabla T - \frac{\bar{\lambda}_c}{c_p} \sum_{p=1}^{n} \frac{1}{c_p} (c_p - c_{p,0}) \nabla Y_i \cdot \nabla T - \frac{n}{c_p} \bar{h}_m.$$

The first term on the RHS of Eq. (7) originates from Fickian diffusion modeling, while the second term inside the parenthesis corresponds to the correction diffusion velocity as suggested by Coffee and Heimerl [16]. For further details concerning the modeling of the diffusion term the reader is referred to Xu et al. [12].

3. Flamelet model

In this section the governing equations are rewritten by considering a reference point that is located within the reaction zone. An asymptotic scaling analysis is then carried out in the vicinity of this reference point leading to a general formulation of the flamelet equations including tangential diffusion effects.

3.1. Governing equations in the vicinity of the reference point

The analysis is carried out for a point of reference $P_b$ which is located in the reaction zone of a curved flame. A Cartesian coordinate system ($x_1, x_2, x_3$) is introduced at this point (see Fig. 3a), which is chosen such that $x_1$ is aligned with the gradient of $Z$ at $P_b$. Consequently, it follows from the definition of the isosurface that first derivatives of $Z$ with respect to $x_3$ are zero ($\beta = 2, 3$), however, second derivatives are non-zero. Since $Z$ is a conserved scalar, it is assumed that there exists a mixture fraction isosurface at the point $P_b$. Its curvature is defined as the negative divergence of the normalized gradient of $Z$ [17] according to Eq. (5).

In the following we consider a projection of the $Z$ isosurface onto the $x_1-x_3$ plane, where $\beta$ is either 2 or 3. This two-dimensional projection is examined in further detail and the following derivation is carried out in the $x_1-x_3$ plane. From Eq. (5) it follows that the overall curvature of the $Z$ isosurface $K_{Z}$ is the sum of the curvatures of the $Z$ isolines $K_{Z,\beta}$ in the $x_1-x_3$ planes

$$K_{Z} = -\nabla \cdot \left( \frac{\nabla Z}{|\nabla Z|} \right) = \sum_{\beta=2} \bar{K}_{Z,\beta},$$

![Fig. 2. Four different conditions for the alignment of isolines of $Z$ and $Y_i$ in the reaction zone of a flame: 2(a) represents the flamelet of an idealized planar counterflow diffusion flame, 2(b) a flamelet of a reacting dispersed particle surrounded by quiescent atmosphere (spherical symmetry), 2(c) the axisymmetric flamelet of a reacting dispersed particle in a convective flow and 2(d) a flamelet in a co-flow flame away from the axis (general case, no symmetry). While the classical flamelet equations are valid for the cases in 2(a) and 2(d), multi-dimensional effects might become relevant for the cases shown in 2(c) and 2(d).](image-url)
is equal to

\[ \kappa_{Z, \beta} = \frac{\rho \frac{\partial \bar{n}}{\partial \bar{n}}}{\rho \frac{\partial n}{\partial n}}. \]

The normal vector of the Z isoline at \( \mathbf{p} \) is defined by

\[ \mathbf{n}_Z = \frac{\nabla Z}{|\nabla Z|} = \frac{1}{\sqrt{\left( \frac{\partial Z}{\partial x} \right)^2 + \left( \frac{\partial Z}{\partial y} \right)^2}} \frac{\begin{pmatrix} \frac{\partial Z}{\partial x} \\ \frac{\partial Z}{\partial y} \end{pmatrix}}{\left| \begin{pmatrix} \frac{\partial Z}{\partial x} \\ \frac{\partial Z}{\partial y} \end{pmatrix} \right|}. \]

A two-dimensional polar coordinate system \((r, \phi)\) is introduced that allows for a convenient description of the Z isoline in the vicinity of \( \mathbf{p} \). The origin of the polar coordinate system is separated from the reference point by

\[ \mathbf{p}_0 = \left( \begin{array}{c} X_{0, r} \\ X_{0, \phi} \end{array} \right) = \mathbf{p} + \frac{1}{\kappa_{Z, \beta}} \mathbf{n}_Z \mathbf{p} = \left( \begin{array}{c} X_{0, r} \\ X_{0, \phi} \end{array} \right). \]

With this, Eq. (3) can be expressed in terms of the two-dimensional polar coordinate system and the gradient and the Laplacian for the general Cartesian coordinate system \((x_1, x_2)\) is given by

\[ \nabla Z = \nabla Z_{1, \beta} = \frac{\partial Z}{\partial x_1} \mathbf{i} + \frac{\partial Z}{\partial x_2} \mathbf{j}. \]

The arc length \( s \) is introduced to measure the distance along the Z isoline, and \( s \) is related to \( \phi \) by

\[ ds = \frac{d\phi}{\kappa_{Z, \beta}}. \]

Since \( s \) is measured along the Z isoline, all derivatives of Z with respect to the arc length are zero. With this, Eq. (16) can be rewritten as:

\[ \frac{\partial Z}{\partial t} + \rho u \frac{\partial Z}{\partial x} + \rho v \frac{\partial Z}{\partial y} - \kappa_{Z, \beta} \rho D_{Z, \beta} \frac{\partial Z}{\partial t} = \frac{\partial}{\partial t} \left( \rho D_{Z, \beta} \frac{\partial Z}{\partial t} \right). \]

In this context, it is noted that the same expression can be obtained by rearranging the diffusion term in Eq. (16) using Eq. (5) [4]:

\[ \nabla^2 Z = \frac{\partial^2 Z}{\partial x^2} + \frac{\partial^2 Z}{\partial y^2} = \frac{\partial^2 Z}{\partial x^2} - \kappa_{Z, \beta} \left[ \frac{\partial Z}{\partial t} \right]. \]

Similarly, equations for species conservation and temperature can be expressed in terms of the coordinate system \((r, \phi)\). Note that derivatives of \( Y_i \) and \( T \) with respect to \( s \) are non-zero. This is a consequence of the fact that the isolines of \( Y_i \) and \( T \) are not necessarily aligned with the isoline of the mixture fraction, which is illustrated in Fig. 2c and d.

Deriving a formulation that is applicable for the general case the equations have to be formulated in a three-dimensional sense. Formally, the previous considerations apply to the \( x_1-x_2 \) plane and the \( x_1-x_3 \) plane, respectively. However, in both reference frames, transport in the flame-normal direction is accounted for. This is readily shown from Eq. (14) by setting \( \phi = 0 \) at \( \mathbf{p}_0 \), thus

\[ dr_{12} = dr_{13} = dx_1. \]

where the indices 12 and 13 correspond to quantities in the \( x_1-x_2 \) plane and in the \( x_1-x_3 \) plane, respectively. Further, two coordinates \( s_p \), describing the local mixture fraction isoline, have to be considered. Consequently, there exist two curvatures, that can be combined to the overall curvature according to Eq. (9). The generalization corresponds to describing the mixture fraction isoline through an ellipsoidal shape in the vicinity of \( \mathbf{p}_0 \). The generalized, three-dimensional equations are obtained by adding the tangential terms, such that the equations for species mass fractions and temperature read

\[ \rho \frac{\partial Y_i}{\partial t} + \rho u \frac{\partial Y_i}{\partial x} + \rho v \frac{\partial Y_i}{\partial y} + \rho D_{Y_i} \frac{\partial^2 Y_i}{\partial x^2} + \rho D_{Y_i} \frac{\partial^2 Y_i}{\partial y^2} = \frac{\partial}{\partial x} \left( \rho Y_i \frac{\partial v}{\partial x} \right) + \frac{\partial}{\partial y} \left( \rho Y_i \frac{\partial v}{\partial y} \right) \]

\[ + \sum_{j=1}^{n_{spec}} \frac{\partial}{\partial y} \left( \rho D_{Y_j} \frac{\partial Y_j}{\partial x} \right) + \frac{\partial}{\partial x} \left( \rho D_{Y_j} \frac{\partial Y_j}{\partial y} \right) \]

\[ + \sum_{j=1}^{n_{spec}} \frac{\partial}{\partial y} \left( \rho D_{Y_j} \frac{\partial Y_j}{\partial x} \right) + \frac{\partial}{\partial x} \left( \rho D_{Y_j} \frac{\partial Y_j}{\partial y} \right) + \rho D_{Y_i} \sum_{j=1}^{n_{spec}} \frac{\partial Y_j}{\partial s_p} \frac{\partial Y_j}{\partial s_p} + \frac{\partial Y_j}{\partial s_p} \frac{\partial Y_j}{\partial s_p}. \]

To facilitate the asymptotic scaling analysis in the next section, Eqs. (21) and (22) are non-dimensionalized by following the following relations:

\[ r^* = \frac{r}{l_m}, \quad \phi^* = \frac{\phi}{l_m}, \quad \rho^* = \frac{\rho}{\rho_x}, \quad D_{Y_i}^* = \frac{D_{Y_i}}{D_{Y_i, st}}, \quad \lambda^* = \frac{\lambda}{l_{at}}, \quad C^*_p = \frac{C_p}{C_p, st}, \]

\[ u^*_a = \frac{u^*_a}{l_m}, \quad m^*_a = \frac{m^*_a}{l_m}, \quad k = \frac{k}{l_m}, \quad t^* = \frac{t}{l_{at}}. \]
and regions 

where \( \Delta Z = 1 \). Since the thermodynamic properties \( \rho, D, \dot{\lambda} \) and \( c_p \) are non-dimensionalized with respect to the thermodynamic state at stoichiometric conditions, they are of order unity in the vicinity of \( P_p \). The species mass fraction \( Y_i \) and the temperature \( T \) are non-dimensionalized with the maximum species mass fraction and the maximum temperature observed in the reaction zone. Thus, \( Y_i \) \( \dot{\lambda} \) and \( T \) vary between zero and unity in the reaction zone. Further, the non-dimensionalized velocity \( v'_r \) is equivalent to the local Peclet number.

3.2. Multi-scale asymptotic scaling and regime analysis

3.2.1. Characteristic scales

An asymptotic scaling analysis is carried out in the vicinity of \( P_p \). It is assumed that the characteristic chemical time scale is short in comparison to convective and diffusive time scales. Consequently, the reaction zone is thin. With this, a short length scale \( l_r \) is introduced:

\[
l_r = \frac{\Delta Z}{V} \frac{Z}{Z_p}, \tag{24}
\]

where \( \Delta Z \) denotes the reaction zone thickness in mixture fraction space. In the following, \( l_r \) is considered as an asymptotic measure for the flame thickness. Being a function of the scalar dissipation rate, the orientation of this length scale is perpendicular to the mixture fraction iso-surface. The correct scaling of this parameter is crucial to the analysis and depends greatly on the assessment of the reaction zone thickness (see discussion in Section 4.1.1).

By considering Eqs. (24) and (25), a small parameter \( \varepsilon \) is defined on the basis of the thin flame sheet assumption

\[
\varepsilon = \frac{\Delta Z}{V} = \frac{l_r}{l_m}. \tag{26}
\]

In addition to the length scale \( l_r \) (flame-normal orientation), corresponding scales in flame-tangential direction are defined as

\[
l_{s, \beta} = \frac{1}{|\kappa_{Z, \beta}|}, \quad \beta = 2, 3. \tag{27}
\]

Assuming that the flame sheet is strongly curved, the two curvatures \( \kappa_{Z, \beta} \) are large, from which two small parameters \( v_{\beta} \) can be identified

\[
v_{\beta} = \frac{l_{s, \beta}}{l_m} \approx \frac{1}{v_{\beta}}. \tag{28}
\]

With the definition of these characteristic scales, we will next perform an analysis assessing the scaling of different terms that appear in the non-dimensionalized Eqs. (21) and (22).

The curvature \( \kappa_{Z, \beta} \) is

\[
\kappa_{Z, \beta} = \kappa_{Z, \beta} l_m = \frac{l_m}{l_{s, \beta}} \approx \mathcal{O}\left(\frac{1}{v_{\beta}}\right). \tag{29}
\]

From Eqs. (23) and (24) it can be readily shown that the gradient of \( Z \) with respect to \( r \) is of order unity in the vicinity of \( P_p \)

\[
\frac{\partial Z}{\partial r} = l_m \frac{\partial Z}{\partial r} = \frac{|\nabla Z|}{|V|} \approx \mathcal{O}(1). \tag{30}
\]

According to Eqs. (17) and (28), the coordinate \( s^r \) is scaled as

\[
s^r = \frac{s^r}{l_m} \approx \mathcal{O}(v_{\beta}). \tag{31}
\]

Using Eq. (26) a short-range coordinate is introduced along the flame-normal direction

\[
\zeta = \frac{Z(r, t) - Z_2}{\varepsilon}. \tag{32}
\]

In the vicinity of \( P_p \) this short-range coordinate is of order unity. The coordinate \( s \), which is measured along the \( Z \) isoline, is rescaled such that it becomes of order unity

\[
\eta_s = \frac{s^r}{v_{\beta}}, \quad \beta = 2, 3. \tag{33}
\]

Since \( v_{\beta} \) is a small parameter due to the large curvature assumption, \( \eta_s \) is a short-range coordinate in that limit.

When moving further away from the point of reference, the short-range coordinates take large values. Thus, derivatives with respect to \( \zeta \) and \( \eta_s \) become negligibly small and the long-range coordinates \( \zeta_1 \) and \( \zeta_2 \), that represent the original coordinates, are used to describe inert mixing and flow field

\[
\zeta_1 = r, \quad \zeta_2 = s, \quad \beta = 2, 3. \tag{34}
\]

From a physical point of view, temporal changes and chemistry play important roles in the vicinity of the flame and have to be accounted for. Consequently, the chemical source term and the transient term may not be dropped in the subsequent analysis of equations for temperature and species mass fractions. Therefore, a new time coordinate is introduced as

\[
\tau = t. \tag{35}
\]

In the following analysis, the chemical source term and terms containing derivatives with respect to \( \tau \) will be treated as leading order terms. The species mass fraction equations are analyzed in detail. All steps of the analysis are analogously carried out for the temperature equation. In the following, asterisks are omitted for notational convenience.

The mass fraction of species \( i \) is expressed as a function of the newly introduced coordinates

\[
Y_i = Y_i(\zeta(Z(r, t)), \eta_s(s), \zeta_1, \zeta_2, \tau). \tag{36}
\]

The non-dimensionalized Eq. (21) is transformed to the coordinate system \( (\zeta_1, \eta_s, \zeta, \zeta_2, \tau) \) by applying the following transformation rules:

\[
\frac{\partial}{\partial \tau} = \frac{\partial}{\partial \tau} + \frac{1}{\varepsilon} \frac{\partial Z}{\partial r} \frac{\partial}{\partial \zeta}, \tag{37}
\]

\[
\frac{\partial}{\partial \zeta} = \frac{1}{\varepsilon} \frac{\partial Z}{\partial r} \frac{\partial}{\partial \zeta} + \frac{\partial}{\partial \zeta_1}, \tag{38}
\]

\[
\frac{\partial}{\partial \zeta_1} = \frac{1}{v_2} \frac{\partial Z}{\partial r_2} + \frac{\partial}{\partial \zeta_2} \frac{\partial}{\partial \zeta_1} + \frac{\partial}{\partial \zeta_2}. \tag{39}
\]

This transformation follows the strategy originally reported in Peters [19, see Eq. 3.128], which was also done for multiple mixture fractions by Hasse and Peters [20]. Rapid changes that originate from reaction–diffusion interaction take place on the short range (coordinates \( \zeta \) and \( \eta_s \)), while slower processes, such as inert mixing and convective processes, take place on the long range (coordinates \( \zeta_1 \) and \( \zeta_2 \)). However, the latter are of negligible influence to the dynamics of the reaction zone. Consequently, terms with respect to \( \zeta \) are of higher order in the vicinity of the point of reference \( P_p \) and can be omitted. The interested reader may follow the transformation term by term in Appendix B.

The scaling of leading order terms appearing in the transformed equations is assessed next. In the following, the use of \( Y_i^s \) and \( Y_i^f \) indicates potential differences for the change of species mass fractions in the flame-normal and the flame-tangential direction, respectively. This notation is kept in all formulations to ensure generality.

The normalized species mass fraction \( Y_i \) is assumed to change substantially on the scale of \( \zeta \) (along flame-normal direction).
Therefore, derivatives of $Y_i$ with respect to $\zeta$ are assumed to be of order unity according to
\begin{equation}
\frac{\partial Y_i}{\partial \zeta} \approx O\left(\frac{\Delta Y_i}{\Delta \zeta}\right) = O(1),
\end{equation}
\begin{equation}
\frac{\partial^2 Y_i}{\partial \zeta^2} \approx O\left(\frac{\Delta Y_i}{\Delta \zeta^2}\right) = O(1).
\end{equation}
This scaling is indicated by the notation:
\begin{equation}
\frac{\partial Y_i}{\partial \eta} \approx \frac{\partial Y_i}{\partial \eta_{fl}} \approx O(\varepsilon),
\end{equation}
where the superscript $\perp$ denotes a quantity that changes in the flame-normal direction.

The quantity $Y_i$ might change less along the flame-tangential direction, since the changing scalar dissipation rate along the $Z$ iso- 
surface might not substantially change neighboring flamelet solutions. Keeping in mind the physical considerations in Section 2, the flame sheet becomes infinitely thin for infinitely fast chemistry ($\varepsilon \to 0$) and the flame is not affected by a changing scalar dissipation rate along the isosurfaces. Thus, no reaction–diffusion interactions occur and gradients in tangential direction vanish. From this rationale, it is concluded that $Y_i$ scales according to $\varepsilon^n$ in flame-tangential direction, where the exponent $n$ is positive. For simplicity, $n$ is set to 1 and the justification of this scaling is examined in Section 4. Thus, derivatives of $Y_i$ with respect to $\eta_{fl}$ scale according to:
\begin{equation}
\frac{\partial Y_i}{\partial \eta_{fl}} = \frac{\partial Y_i}{\partial \eta_{fl}} \approx O(\varepsilon).
\end{equation}
\begin{equation}
\frac{\partial^2 Y_i}{\partial \eta_{fl}^2} \approx O\left(\frac{\Delta Y_i}{\Delta \eta_{fl}}\right) = O(\varepsilon).
\end{equation}

To describe that scaling within the transformed species equation, a rescaled quantity $Y_i' = Y_i/\varepsilon$ of order unity is introduced. Thus, species gradients in flame-tangential direction are rescaled to order unity
\begin{equation}
\frac{\partial Y_i'}{\partial \eta_{fl}} = \frac{\partial Y_i}{\partial \eta_{fl}} \approx O(1).
\end{equation}

As a result, derivatives of $Y_i$ with respect to $s_{fl}$ scale as
\begin{equation}
\frac{\partial Y_i}{\partial s_{fl}} = \frac{\partial Y_i}{\partial \eta_{fl}} \approx O\left(\frac{\Delta Y_i}{\Delta \eta_{fl}}\right) \approx O\left(\frac{\varepsilon}{v'}\right),
\end{equation}
\begin{equation}
\frac{\partial^2 Y_i}{\partial s_{fl}^2} \approx O\left(\frac{\varepsilon}{v' \varepsilon}\right) \approx O\left(\frac{\varepsilon}{v'}\right)^2 \approx O(1).
\end{equation}

This scaling is also consistent from a physical perspective since gradients with respect to $s_{fl}$ are promoted by large curvatures (see definition of $v'$, Eq. (28)) but vanish in the limit of infinitely fast chemistry ($\varepsilon \to 0$).

Due to the fact that $\rho$ and $D_i$ are proportional to $T^{-1}$ and $T^n$, respectively, and keeping in mind that the previous relations are analogous valid for the temperature, derivatives of $\rho D_i$ are rescaled in tangential direction
\begin{equation}
\frac{\partial (\rho D_i)}{\partial \eta_{fl}} = \frac{\partial (\rho D_i)}{\partial \eta_{fl}} \approx O(1).
\end{equation}
\begin{equation}
\frac{\partial^2 (\rho D_i)}{\partial \eta_{fl}^2} \approx O\left(\frac{\varepsilon}{v' \varepsilon}\right) \approx O\left(\frac{\varepsilon}{v'}\right)^2 \approx O(1).
\end{equation}
The scaling $\varepsilon^{-1}$ in the above equation is a conservative measure but simplifies the further analysis. In reality the exponent is $\alpha - 1$ according to the previous considerations and this term takes values between 0 and $-1$. Furthermore, rescaled curvatures of order unity are introduced
\begin{equation}
K_{fl} = \frac{v'}{\varepsilon} K_{\perp} \approx O(1).
\end{equation}

Another small parameter $v$ can be introduced using the curvature defined by Eq. (28)
\begin{equation}
v = \frac{L_k}{l_m} = \frac{1}{|K_2|l_m}.
\end{equation}

Simplifying the transformed terms it can be shown that
\begin{equation}
\sum_{j=1}^{m} \frac{1}{v' v} \approx O\left(\frac{1}{v^2}\right),
\end{equation}
\begin{equation}
O\left(\frac{1}{v^2}\right) \approx O\left(\frac{1}{v^2}\right). \quad (51)
\end{equation}

where Eqs. (9) and (28) are used. Collecting all leading order terms after the transformation of Eq. (21) yields
\begin{equation}
\rho \frac{\partial Y_i}{\partial t} + \sum_{j=2}^{3} \rho v_{y_j} \frac{\partial Y_i}{\partial y_j} - \frac{1}{v'^2} \left[\frac{\varepsilon}{4} \left(\frac{Le_i}{1} - 1\right) \frac{\partial (\rho D_i)}{\partial \eta_{fl}} + \frac{1}{v'^2} \rho D_i \Delta Y_i \right]
\end{equation}
\begin{equation}
+ \frac{1}{v'^2} \left[\frac{\varepsilon}{4} \left(\frac{Le_i}{1} - 1\right) \frac{\partial (\rho D_i)}{\partial \eta_{fl}} + \frac{1}{v'^2} \rho D_i \Delta Y_i \right]
\end{equation}
\begin{equation}
- \frac{1}{v'^2} \left[\frac{\varepsilon}{v} \rho D_i \Delta Y_i \right] \approx 0. \quad (52)
\end{equation}

The detailed derivation of Eq. (52) is given in Appendix B. To maintain generality, the Lewis number of the mixture fraction is kept variable, although it is set to unity in Eq. (3). Due to the similarity of the equations, a notation analogous to Xu et al. [12] is chosen. In their work, the origin of the individual terms is extensively discussed. Differences between the equations are found in the terms $f_{c_1}$ and $f_{c_2}$, which simplify for an axial flamelet as investigated in [12] (see Appendix C). Additionally, the term $f_{c_3}$ originates from variable Lewis numbers, which are set constant in [12]. With respect to the previous definitions, the term $f_{c_1}$ represents multi-dimensional transport along mixture fraction isosurfaces and $f_{c_2}$ originates from the diffusion correction of $f_{c_1}$. The terms $f_{c_3}$ and $f_{c_4}$ represent differential diffusion in flame-tangential direction and the corresponding diffusion correction, respectively.

3.2.2. Consideration for unity Lewis numbers
For the condition of unity Lewis numbers, all terms except for $f_{c_1}$, $f_{c_2}$, and the chemical source term vanish on the RHS of Eq. (52)
\begin{equation}
\rho \frac{\partial Y_i}{\partial t} + \sum_{j=2}^{3} \rho v_{y_j} \frac{\partial Y_i}{\partial y_j} - \frac{1}{v'^2} \left[\frac{\varepsilon}{4} \left(\frac{Le_i}{1} - 1\right) \frac{\partial (\rho D_i)}{\partial \eta_{fl}} + \frac{1}{v'^2} \rho D_i \Delta Y_i \right] \approx 0. \quad (53)
\end{equation}

Thus, the leading order terms are either of order $1/\varepsilon^2$ or $\varepsilon/v^2$. Using the standard flamelet term $f_{c_1}$ as reference, the equation is multiplied by $\varepsilon^2$ such that the prefactor of $f_{c_1}$ cancels. The remainder of this term is of order unity by the previous definitions. From the prefactor that appears for the multi-dimensional term $f_{c_1}$ it is
concluded that there exists a characteristic ratio between $\varepsilon$ and $\nu$ that reads

$$\frac{\varepsilon^3}{\nu^2} = \frac{(\Delta Z)_k^2}{\nu Z_0^2} = \frac{2(\Delta Z)_k^2 D_Z}{\nu D_p}. \quad (54)$$

From the size of that characteristic ratio the relevance of multi-dimensional effects can be assessed. In case it is order unity, $f_{c_1}$, is of the same order of magnitude as $f_{c_2}$. On the other hand, if the ratio becomes smaller or greater than unity, either $f_{c_1}$ or $f_{c_2}$ are of leading order, respectively.

The important characteristics that have been deduced from physical considerations in Section 2 are well reflected by this characteristic ratio. In the limit of infinitely fast chemistry, the reaction zone thickness in mixture fraction space $(\Delta Z)_k$ tends to zero and the classical flamelet equations are recovered ($f_{c_1}$ vanishes in Eq. (53)). Further, assuming a planar flame, thus $(\Delta Z)_k \to 0$, also leads to the classical flamelet formulation. Therefore, this ratio leads to two distinguished limits that are shown in Table 1. These limits determine whether multi-dimensional effects have to be considered in the flamelet equations and can be interpreted as regimes. Since for the first regime the classical flamelet equations as derived by Peters [1] are valid it is called the classical flamelet regime. On the other hand, for strongly curved flames at finite rate chemistry the characteristic ratio can become greater than one. In that case, multi-dimensional effects become relevant in Eq. (53). Consequently, the corresponding regime is denoted as multi-dimensional flamelet regime.

3.2.3. Consideration for non-unity Lewis numbers

For the consideration of differential diffusion effects, it can be seen from Eq. (52) that leading order terms appear to be either of order $1/\varepsilon^2$, $(Le_2/Le_1 - 1)/\varepsilon^2$ or $\varepsilon^2/\nu^2$. The relevance of the terms $f_{c_1}$ and $f_{c_2}$ representing transport along mixture fraction isosurfaces, is assessed by the previously discussed distinguished limits. Furthermore, for the term $f_{c_3}$ another characteristic ratio is obtained analogously to Section 3.2.2:

$$\frac{\varepsilon}{\nu} \left( \frac{Le_2}{Le_1} - 1 \right) = \left( \frac{\Delta Z}_{a} \right) \left| \frac{2D_Z}{D_p} \right| \left( \frac{Le_2}{Le_1} - 1 \right). \quad (55)$$

Similarly to the characteristic ratio in Eq. (54) it shows that $f_{c_2}$ will vanish for planar flames $(\Delta Z)_k \to 0$ or infinitely fast chemistry $(\Delta Z)_k \to 0$. Furthermore the term becomes negligible for species with $Le_1 \to Le_2$, because $(Le_2/Le_1 - 1)$ tends to zero. However, assuming there exist species with Lewis numbers that deviate significantly from $Le_2$, the term $(Le_2/Le_1 - 1)$ becomes order unity. Since we are interested in a measure when $f_{c_3}$ becomes relevant, it is reasonable to simplify the characteristic ratio to $\varepsilon/\nu$ in Eq. (55). This ratio is also obtained for the term $f_{c_4}$ that originates from the diffusion correction of $f_{c_2}$.

Using both characteristic ratios, three regimes can be identified that determine the relevance of tangential diffusion terms in the flamelet equations. These regimes correspond to deriving multiple distinguished limits when considering non-unity Lewis numbers. In addition to the previously discussed regimes, the classical flamelet regime (regime I) and the multi-dimensional flamelet regime (regime III), respectively, there is an intermediate regime II. In that regime, differential diffusion along flame-tangential direction

<table>
<thead>
<tr>
<th>Regime name</th>
<th>Regime description</th>
<th>Characteristic ratio</th>
<th>Distinguished limit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Classical flamelet regime</td>
<td>$e^3 &lt; \nu^2$</td>
<td>$2(\Delta Z)_k^2 D_Z/\nu Z_0$</td>
<td>$\varepsilon &lt; \nu$</td>
</tr>
<tr>
<td>Multi-dimensional flamelet regime</td>
<td>$e^3 &gt; \nu^2$</td>
<td>$2(\Delta Z)_k^2 D_Z/\nu Z_0$</td>
<td>$\varepsilon &gt; \nu, \nu^3 &gt; e^2$</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Regime name</th>
<th>Tangential diffusion effects</th>
<th>Distinguished limit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Regime I Classical flamelet regime</td>
<td>None</td>
<td>$e &lt; \nu$</td>
</tr>
<tr>
<td>Regime II Curvature-affected flamelet regime</td>
<td>$f_{c_3}$ and $f_{c_4}$</td>
<td>$\varepsilon &gt; \nu, \nu^3 &gt; e^2$</td>
</tr>
<tr>
<td>Regime III Multi-dimensional flamelet regime</td>
<td>$f_{c_1} - f_{c_2}$</td>
<td>$\varepsilon &gt; \nu, \nu^3 &gt; e^2$</td>
</tr>
</tbody>
</table>

Table 2: The regimes derived from asymptotic scaling analysis for $Le 
eq 1$.

In addition to the previously discussed regimes, the classical flamelet equations are recovered ($f_{c_1}$ vanishes in Eq. (53)). Further, assuming a planar flame, thus $(\Delta Z)_k \to 0$, also leads to the classical flamelet formulation. Therefore, this ratio leads to two distinguished limits that are shown in Table 1. These limits determine whether multi-dimensional effects have to be considered in the flamelet equations and can be interpreted as regimes. Since for the first regime the classical flamelet equations as derived by Peters [1] are valid it is called the classical flamelet regime. On the other hand, for strongly curved flames at finite rate chemistry the characteristic ratio can become greater than one. In that case, multi-dimensional effects become relevant in Eq. (53). Consequently, the corresponding regime is denoted as multi-dimensional flamelet regime.

3.3. Generalized flamelet equations

In this section, we summarize the general form of the flamelet equations for species mass fractions and temperature. These equations are obtained from Eq. (52) using the transformation $dZ = ed\varepsilon$ and $ds_p = vfd\eta$:

$$\rho \frac{\partial Y_i}{\partial \tau} + \sum_{k=2}^{n} \rho v_i \frac{\partial Y_i}{\partial \eta} - Le 2 \rho Y_i \frac{\partial^2 Y_i}{\partial \eta^2} + \frac{1}{4} \left( \frac{Le_2}{Le_1} - 1 \right) \frac{\partial(\rho Y_i)}{\partial \eta} \frac{\partial Y_i}{\partial \eta}$$

$$+ \frac{1}{4} \left( \frac{Le_2}{Le_1} - 1 \right) \frac{\partial(\rho D_Y)}{\partial \eta} \frac{\partial Y_i}{\partial \eta}$$

$$- \frac{\rho D_Y n Y_i \sum_{k=1}^{m} Le_k \frac{\partial Y_k}{\partial \eta}}{4 \sum_{k=1}^{m} Le_k \frac{\partial Y_k}{\partial \eta}} + \frac{\rho Y_i \frac{\partial Y_i}{\partial \eta}}{2 \sum_{k=1}^{m} Le_k \frac{\partial Y_k}{\partial \eta}}$$

$$+ \rho D_Y \sum_{k=1}^{m} \frac{\partial^2 Y_k}{\partial \eta^2} \frac{Le_k}{Le_2} \frac{\partial Y_k}{\partial \eta} + m_i, \quad (56)$$

and the flamelet equation for temperature reads (for the detailed derivation see Appendix B):

$$\frac{\partial T}{\partial \tau} + \sum_{k=2}^{n} \rho v_i \frac{\partial T}{\partial \eta} - Le 2 \rho Y_i \frac{\partial^2 T}{\partial \eta^2} + \frac{1}{2} \left( \frac{Le_2}{Le_1} - 1 \right) \frac{\partial(\rho Y_i)}{\partial \eta} \frac{\partial Y_i}{\partial \eta}$$

$$+ \frac{1}{2} \left( \frac{Le_2}{Le_1} - 1 \right) \frac{\partial(\rho D_Y)}{\partial \eta} \frac{\partial Y_i}{\partial \eta}$$

$$+ \frac{1}{4} \left[ \frac{\rho Y_i \frac{\partial Y_i}{\partial \eta}}{2 \sum_{k=1}^{m} Le_k \frac{\partial Y_k}{\partial \eta}} \right]$$

$$+ \rho D_Y \sum_{k=1}^{m} \frac{\partial^2 T}{\partial \eta^2} \frac{Le_k}{Le_2} \frac{\partial Y_k}{\partial \eta} + m_i, \quad (57)$$

and the flamelet equation for temperature reads (for the detailed derivation see Appendix B):
It can be shown that Eqs. (56) and (57) are consistent with previous works [4,12,13,21,22] (the interested reader is referred to Appendix D for term-by-term comparisons). For instance, a similar set of coordinates was used by Williams [21], with Z as the normal coordinate and two variables x and y that span the mixture fraction iso-surface. Analogously, Williams [21] obtained flamelet equations including terms that account for normal and tangential transport. Kortchik et al. [4] were the first to introduce curvature-based correction terms into flamelet equations similar to the term ftc. Following the notation of Xu et al. [12] in this work, Eqs. (56) and (57) can be readily related to their flamelet equations. However, they introduced simplifications due to symmetry such that their multi-dimensional terms represent a simplified form of the corresponding terms in this work. In summary, the subject of interest is clearly similar in these previous works. The researchers have seemingly introduced their own set of notations to describe similar contributions and they frequently focused on certain aspects of tangential diffusion processes. Therefore, it may be worthwhile to adopt a general form of the flamelet equations, such as that shown in Eqs. (56) and (57), which will naturally account for all relevant effects from which simplifications follow in a natural way. Furthermore, the asymptotic scaling analysis and regime classification, derived in this work, may be used to assess the importance of tangential diffusion processes relative to normal diffusion processes.

The asymptotic scaling analysis was carried out at a reference point P0 and it was shown that under certain conditions the one-dimensional structure of the flamelet equations is violated. This is the case when the point of reference is situated in regime III (see Tables 1 and 2), where multi-dimensional effects (terms ftc and ftg) become relevant. These effects result from variations of the scalar dissipation rate along iso-surfaces of Z. Since χ is dependent on Z, it is assumed that structures based on Z can still be utilized to solve the equations. Thus, flamelets are reconstructed from the trajectories of the mixture fraction gradient in Section 4.

Multi-dimensional effects are interpreted as transport between neighboring flamelets. Note that differential diffusion in flame-tangential direction (terms ftc and ftg) are not multi-dimensional. In regime II, where these effects are expected, a one-dimensional flamelet structure can be retained. It was shown by Xuan et al. [13] that considering only differential diffusion in tangential direction and neglecting multi-dimensional terms already leads to improved results.

In the following, the curvature and the scalar dissipation rate are treated as functions of Z since both quantities might change when moving along a flamelet. With K2 and χ being functions of Z, hence, the characteristic ratio is dependent on Z. Consequently, flamelets might undergo a transition from one regime to another which is discussed in Section 4.

4. Results

In this section we examine a laminar and a turbulent flame to verify the asymptotic scaling analysis using numerical results. Since the relevance of diffusion effects in flame-tangential direction has been reported for laminar jet flames [8,13], we review a laminar methane–air co-flow flame [10] first. Thereafter, we investigate a turbulent lifted hydrogen jet flame [15] to determine the relevance of tangential diffusion for turbulent flames.

4.1. Laminar co-axial jet diffusion flame

The co-flow flame originally studied by Bennett et al. [10] is well documented in the literature and has been investigated experimentally and numerically [5–7,9,10,23]. A fuel stream is introduced through a tube (inner jet) while a co-flowing air stream encloses the fuel stream (outer jet). Bennett et al. [10] studied partially premixed and non-premixed flame configurations (different equivalence ratios Φ in the inner jet), however, only the non-premixed flame configuration (Φ = ∞) is considered here. For further details on the flame configuration the reader is referred to Bennett et al. [10].

The CFD solution method was described in detail by Messig et al. [9]. A two-dimensional axisymmetric block mesh with a total of 185,000 cells (refined near the axis) is used for the computations. This mesh resolution ensures that the flamelet budgets discussed below are fully consistent with the CFD solution. For the detailed chemistry solution the GRI 1.2 mechanism [24], consisting of 32 species and 177 reactions, is applied. The mixture fraction is treated as a conserved scalar and computed from Eq. (3) setting its Lewis number Le = 1. The Lewis numbers for all species are set either to unity or to constant, but non-unity values. Both cases will be discussed in the following. The values for the non-unity Lewis numbers are given in Appendix A. All relevant terms for the budget analysis were post-processed from the scalar fields of the numerical solutions.

Figure 4 shows mixture fraction and CO2 mass fraction isolines at the top and the temperature contours at the bottom. Consistent with the discussion in Section 2, it can be seen that the scalar fields of Z and YCO2 do align outside the reaction zone while the isolines become misaligned in the reaction zone. Further, it is noted that the scalar fields exhibit the largest curvatures near the axis. According to the asymptotic scaling analysis high curvature promotes multi-dimensional effects. Therefore, three different flamelets, shown in Fig. 4, are extracted for a budget analysis corresponding to a flamelet along the centerline (F1), a location near the flame tip (F2) and a flamelet solution at the flame-base (F3). These flamelets were reconstructed from trajectories of the mixture fraction gradient. Thereafter, the terms of Eqs. (56) and (57) are evaluated along these flamelets for cases with Le = 1 and Le=1.

The reaction zone is located in the vicinity of the maximum temperature with respect to each flamelet. For the flamelets F1, F2 and F3 the maximum temperature is found at Z = 0.057, 0.059 and 0.063, respectively (case Le = 1). Therefore, the region 0.05 < Z < 0.07 is of particular interest for the analysis and is denoted as reaction zone in the following.

![Fig. 4. Simulations of a laminar co-axial jet flame, showing top: the Z isolines (black) and the YCO2 isolines (red) and bottom: the temperature distribution. The thick black line marks Zε = 0.055. Furthermore, the flamelets (solid blue lines) F1 (located on the flame centerline), F2 and F3 are displayed. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)](image-url)
4.1.1. Analysis of scaling relations

The scaling relations are examined in detail in this section. An estimate for \((\Delta Z)_k\) is found in the book by Peters [19, see page 217] and in the work of Seshadri and Peters [25]. The latter analyzed the flame structure of non-premixed methane–air counterflow flames. In their works, the overall reaction zone thickness is determined by the largest layer, which is called the oxidation layer. With respect to the rather small scalar dissipation rates that are determined by the largest layer, which is called the oxidation layer. In their works, the overall reaction zone thickness is determined by the largest layer, which is called the oxidation layer. In their works, the overall reaction zone thickness is determined by the largest layer, which is called the oxidation layer.

In this work, the small parameter \(\varepsilon\) is assessed for each flamelet using Eq. (41) and the relation \(d\varepsilon = \partial Z\varepsilon\):  
\[
\varepsilon = (\Delta Z)_k = \sqrt{Y_{i,max}\max\left(\frac{\partial Y_i}{\partial Z}\right)_k},
\]  
\[(58)\]

where \(\max(\cdot)_k\) indicates the maximum in the reaction zone. Note that Eq. (58) applies to the non-dimensionalized derivative \(\partial^2 Y_i / \partial Z^2\). As the reference quantity, CO is chosen as species \(i\) for the assessment of \(\varepsilon\). It is emphasized that this choice is not arbitrary and that \(\varepsilon\) serves as an approximate scale for a single flamelet. If CH$_4$ was chosen as the reference species, flame-normal transport terms in the species equation of CO would be overpredicted leading to an erroneous regime classification. This is due to the fact, that CH$_4$ is consumed in the fuel consumption layer, which is an order of magnitude smaller than the overall flame thickness [25]. To capture all influences of tangential diffusion on the overall flame structure, a rather conservative measure for \(\varepsilon\) is sought. This is achieved by choosing a quantity that changes on the scale of the overall flame thickness, such as CO or the temperature.

Evaluating Eq. (58) for the flamelets F1, F2 and F3 yields values in the range of 0.008 to 0.015 for \(\varepsilon\) (and the reaction zone thickness \((\Delta Z)_k\), respectively) as shown in Table 3. Hence, the estimate of the reaction zone thickness in mixture fraction space according to Eq. (58) is in good agreement with the values of Seshadri and Peters [25].

Verifying the assessment of \(\varepsilon\) numerically, the re-scaled, flame-normal derivatives with respect to \(\zeta\) are computed for the flamelets F1, F2 and F3 according to
\[
\frac{\partial Y_i}{\partial \zeta} = \varepsilon \frac{\partial Y_i}{\partial Z},
\]  
\[
\frac{\partial^2 Y_i}{\partial \zeta^2} = \varepsilon^2 \frac{\partial^2 Y_i}{\partial Z^2},
\]  
\[(59)\]

where the major species CO, CO$_2$, H$_2$, H$_2$O and the radical species OH are chosen for the species \(i\). Figure 5 confirms that these derivatives are of order unity for all flamelets as imposed by the asymptotic scaling analysis.

Using the previously determined scaling parameter \(\varepsilon\), the exponent \(n\), discussed above Eq. (43), is examined next. This exponent is relevant for the assessment of multidimensional effects (scaling of \(f_{c1}\) in Eq. (52), analogously valid for \(ftc\) in Eq. (57)). Keeping in mind that the term \(f_{c1}\) scales with \(\varepsilon^n / \varepsilon^2\) (this can be readily shown when keeping \(n\) in Eqs. (43) and (44) in the subsequent analysis), \(n\) can be computed for each flamelet according to
\[
n = \log\left(\frac{\varepsilon^2 / f_{c1}}{\log(\varepsilon)}\right).
\]  
\[(61)\]

Figure 6 shows that \(n\) varies between 1.1 and 2.3 in the reaction zone of the flamelets F1, F2 and F3 for the species CO, CO$_2$, H$_2$ and H$_2$O. However, for general applicability a constant value for \(n\) is sought. Setting it to a lower bound with \(n = 1\) can be interpreted as a conservative measure to ensure that all relevant multi-dimensional effects in the reaction zone are covered by regime III.
The terms \( f_1 \) and \( f_3 \) scale according to \( \varepsilon/\nu^2 \) and \( (Le/Le_0 - 1)/\nu \), respectively. When divided by their characteristic scales, these terms are re-scaled to order unity. Figure 7 shows that re-scaling for the flamelet \( F_2 \). It is observed that the re-scaled terms are situated below the dashed red line marking \( O(1) \). Consequently, the scaling relations provide a conservative measure discussed above, such that the terms \( f_1 \) and \( f_3 \) are slightly over-predicted throughout the reaction zone. Similar characteristics are found for the flamelets \( F_1 \) and \( F_3 \) (not shown). It is emphasized that the scaling relations could be further improved by adjusting the previously examined exponent \( n \).

In the following section the scaling relations are used together with the budgets to investigate the relevance of tangential diffusion effects in the flamelet equations.

### 4.1.2. Budget analysis for unity Lewis numbers

Results from the budget analysis for the species \( CO, H_2, OH \) and the temperature are shown in Fig. 8 for all three flamelets. From these results it can be observed that multi-dimensional effects (contained in \( f_3 \)) and the chemical source term are dominant along the flamelet \( F_1 \) for all flamelet equations and all other terms are small in comparison. The term \( f_1 \), also denoted as standard flamelet term, shows only small influence throughout the reaction zone for all budgets. This is also consistent with the assumption of a boundary layer flow at the axis where the second derivatives in axial direction \( (\partial^2/\partial Z^2) \) become negligible. Furthermore, Fig. 9 shows the characteristic ratios for the flamelets \( F_1, F_2 \) and \( F_3 \). It is observed that the characteristic ratio \( \nu^2/\nu^3 \) is significantly larger than unity for \( F_1 \) throughout the reaction zone suggesting that regime III applies for that flamelet.

In contrast to flamelet \( F_1 \), the budget analysis for flamelet \( F_3 \) in Fig. 8 indicates that multi-dimensional terms are negligibly small. Comparing these findings to Fig. 9 shows that the characteristic ratio is several orders of magnitude smaller than unity throughout the reaction zone. Consequently, regime I applies to flamelet \( F_3 \). Interestingly, these effects can also be seen in Fig. 4, which shows that the flamelet \( F_3 \) passes only through regions where the isolines of \( Z \) and \( Y_{CO} \) appear to align. In the region where \( F_3 \) passes through the reaction zone (regions of high temperatures, see bottom of Fig. 4) the isolines of the two scalar fields are weakly curved. These observations are consistent with the distinguished limits derived from Eq. (54).

The flamelet \( F_2 \) was chosen as an intermediate between \( F_1 \) and \( F_3 \). From Fig. 4 it is observed that it passes through the reaction zone close to the axis, where the scalar fields of \( Z \) and \( Y_{CO} \) are still strongly curved and not fully aligned. Further away from the axis the isolines of the scalar fields are weakly curved and aligned. This transition from a region where multi-dimensional effects play a role to a region where they are negligible is also reflected in the budget analysis of the flamelet equations (Fig. 8). Between \( Z = 0.3 \) and a transition point close to \( Z_{st} (0.055) \) the term \( f_1 \) is dominant over the standard flamelet term \( f_1 \) in the flamelet species equations. From the transition point towards the oxidizer boundary \( (Z = 0) \), however, the standard term becomes dominant over the multi-dimensional term. This effect is also supported by Fig. 9 where a transition of the characteristic ratio is observed in the reaction zone. Hence, flamelet \( F_2 \) undergoes a transition from regime I to III according to the limits in Table 1. One of the reasons for this transition between regimes is the scalar dissipation rate profile of flamelet \( F_2 \), which is shown in Fig. 10. Since it formally represents a diffusion coefficient for flame-normal transport terms in Eqs. (56) and (57), high scalar dissipation rates promote flame-normal diffusion. In contrast to the \( \chi \)-profile of flamelet \( F_1 \), it is observed that the \( \chi \)-profile for flamelet \( F_2 \) in Fig. 10 shows a second maximum towards the oxidizer side. The characteristic point of inflection of the latter corresponds to the point, where flamelet \( F_2 \) recedes from the symmetry axis in Fig. 4.

### 4.1.3. Budget analysis for non-unity Lewis numbers

For the cases with \( Le \neq 1 \) two characteristic length scale ratios (see Eqs. (54) and (55)) and the corresponding distinguished limits (see Table 2) were derived. These ratios lead to the regime diagram in Fig. 11 as already discussed in Section 3.2.3. The points of the flamelets \( F_1, F_2 \) and \( F_3 \) located in the reaction zone are shown in the regime diagram. Similarly to the case with unity Lewis numbers, the regime diagram suggests that \( F_1 \) is dominated by tangential diffusion effects while standard flamelet terms predominantly govern the flamelet structure for \( F_3 \). Furthermore, the asymptotic scaling analysis predicts a transition between regimes for flamelet \( F_2 \) also in the case with \( Le \neq 1 \).

Budget analyses of the flamelet equations for the case with \( Le \neq 1 \) are shown in Fig. 12 for the flamelets \( F_1 \)–\( F_3 \). In accordance with the predictions of the asymptotic scaling analysis, the flamelet \( F_1 \) exhibits dominant tangential diffusion effects (terms \( f_1-f_3 \)) while standard flamelet terms (\( f_1-f_2 \)) are negligible. This can be observed for all budgets of the temperature and species equations in Fig. 12. Comparing the budgets for flamelet \( F_1 \) for \( Le = 1 \) (see Fig. 8) and \( Le \neq 1 \) (see Fig. 12) it is observed that differ-

<table>
<thead>
<tr>
<th>Flamelet</th>
<th>( \varepsilon ) (( Le = 1 ))</th>
<th>( \varepsilon ) (( Le \neq 1 ))</th>
</tr>
</thead>
<tbody>
<tr>
<td>( F_1 )</td>
<td>0.011</td>
<td>0.013</td>
</tr>
<tr>
<td>( F_2 )</td>
<td>0.011</td>
<td>0.008</td>
</tr>
<tr>
<td>( F_3 )</td>
<td>0.013</td>
<td>0.015</td>
</tr>
</tbody>
</table>
ential diffusion has considerable impact on the flame structure. All budgets show different structures with respect to the spatial influence region, the magnitude and the sign of the individual terms. This emphasizes the importance of differential diffusion when considering tangential diffusion effects in laminar flames, which is in accordance with previous findings [8]. As expected, the term \( f_{c_3} \) shows significant influence for species whose Lewis number deviates significantly from unity, e.g. \( H_2 \) (\( \text{Le}_{H_2} = 0.296 \)). For species with Lewis numbers close to unity, such as \( \text{CO} \) (\( \text{Le}_{\text{CO}} = 1.073 \)), the term \( f_{c_1} \) is small compared to the other terms. Overall, \( f_{c_3} \) shows significant influence for most of the species budgets of flamelet \( F_1 \). Further, it is noted that, besides the chemical source term, \( f_{c_1} \) is of leading order for flamelet \( F_1 \) throughout the reaction zone. Additionally, the tangential diffusion terms \( f_{c_2} \) and \( f_{c_3} \), which originate from the diffusion correction, remain small in all budgets.

Comparing the budgets for flamelet \( F_3 \) in Figs. 8 and 12, it is noted that tangential diffusion is negligible for both cases, \( \text{Le} = 1 \) and \( \text{Le} \neq 1 \), respectively. Instead, standard flamelet terms are dominant, which corresponds to regime I. The flame structure that is assessed from the budgets is only slightly affected by consideration of differential diffusion. However, the overall flame characteristics remain comparable. This is due to the proximity of flamelet \( F_3 \) to the burner nozzle where large gradients (thus, high scalar dissipation rates) and large shear is observed. Both characteristic ratios reflect the trend that increasing \( \chi \) will move the flamelet toward the regime where classical flamelet equations are valid (regime I).

Similar to the case with \( \text{Le} = 1 \), a transition point is observed for the budgets of flamelet \( F_2 \) where tangential diffusion becomes less influential while standard flamelet terms become relevant. This trend is also reflected by the regime diagram in Fig. 11 indicating a transition from regime III to l.

### 4.2. Turbulent lifted hydrogen jet flame

In the following, we will extend the asymptotic scaling analysis to the turbulent flame regime. The configuration of interest is a direct numerical simulation (DNS) of a spatially developing turbulent lifted hydrogen jet flame in a heated coflow. This configuration was studied by Yoo et al. [15]. The DNS was performed at a jet Reynolds number of 8000, based on mean fuel injection velocity of 240 m/s, jet width of 2 mm, and fuel viscosity of \( 6 \times 10^{-5} \text{ m}^2/\text{s} \). The computational domain of the flame is a box with dimensions of \( 30 \times 40 \times 6 \text{ mm} \) in streamwise (x), transverse (y), and spanwise (z) directions, respectively. The domain is discretized by a Cartesian grid of \( 2000 \times 1600 \times 400 \) points in order to fully resolve all scales of the turbulent flow. The reactions were represented by a detailed hydrogen–air chemical kinetic mechanism, which consists of nine species and 21 elementary reactions [26]. The thermodynamic and mixture-averaged transport properties of the species were calculated with the CHEMKIN and TRANSPORT software libraries [27,28], respectively. The subsequent flamelet budget analyses were based on the diagnostics developed in a previous investigation of a reacting hydrogen/air jet-in-cross-flow [29].

An instantaneous planar view of the temperature field along the mid-plane is shown in Fig. 13. The thick black line indicates the isoline of the stoichiometric mixture fraction \( Z_{st} = 0.2 \), and the blue curves correspond to different instantaneous flamelet projections that were extracted from the DNS. Four flamelets, each enclosed by a window in Fig. 13, are selected for the following budget analyses. These four flamelets correspond to different scenarios, and therefore are representative of the entire DNS dataset. For convention, the four flamelets are distinguished by their stoichiometric streamwise locations \( X_{st} = (5, 14, 17, 27) \text{ mm} \) and are labeled 1–4, respectively.

To better illustrate the chosen flamelets, sub-plots showing a local region in the vicinity of each flamelet are provided in Fig. 13, overlaying the contour of the mixture fraction. Similar to the temperature plot, the thick black line in these sub-plots denotes the stoichiometric mixture fraction.

In regard to the relevance of the multi-dimensional effects in turbulent reacting flows, we will first provide a physical interpretation of the effects. By definition, the multi-dimensional terms are responsible for the diffusion of thermochemical quantities along the mixture fraction isosurface. When properly accounted for, these flamelet terms will enable interactions among flamelets. Interestingly, these mechanisms can be illustrated by plotting the
Fig. 8. Budgets of the flamelet equations for the flamelets $F_1$, $F_2$ and $F_3$ for $Le = 1$. The budgets are shown for the interval $0 < Z < 0.25$ where the chemical source term influences the budgets. The value of the stoichiometric mixture fraction $Z_{st}$ is 0.055 (vertical dashed line).

Fig. 9. The characteristic ratio as a function of the mixture fraction $Z$ for the flamelets $F_1$, $F_2$ and $F_3$ for $Le = 1$. Additionally, the limiting line (dashed line) that corresponds to the distinguished limits from Table 1 is shown.

Fig. 10. The scalar dissipation rate profiles throughout the reaction zone for the flamelets $F_1$, $F_2$ and $F_3$ for $Le = 1$. For all three flamelets the profiles exhibit a maximum around $Z = 0.5$ (not shown in the graph). Further, the profiles of the flamelets $F_2$ and $F_3$ exhibit a second maximum towards the oxidizer side.
Fig. 11. The flamelet regime diagram corresponding to the regimes in Table 2. For the case including differential diffusion, case $Le \neq 1$, three regimes are identified (see Fig. 11b). For equal diffusivities ($Le = 1$) regime II vanishes (see Fig. 11a). Points located within the interval $0.05 < Z < 0.07$ are plotted for the flamelets $F_1$, $F_2$ and $F_3$. The dashed black line and the solid black line correspond to the characteristic ratios (see Eqs. (54) and (55), respectively).

Fig. 12. Budgets of the flamelet equations for the flamelets $F_1$, $F_2$ and $F_3$ for $Le \neq 1$. The budgets are shown for the interval $0 < Z < 0.25$ where the chemical source term influences the budgets. The value of the stoichiometric mixture fraction $Z_{st}$ is 0.055 (vertical dashed line).
multi-dimensional terms as field variables; this is illustrated in Fig. 14. The contour on the left plot shows the $ftc$-term, which is overlapped by several temperature contour lines in the range of $[1300, 2100]$ K. The right plot shows the $ftc_2$-profile along with the $Z = [0.1, 0.4]$ isolines. Both profiles have been non-dimensionalized by the instantaneous planar-average heat-release rate, corresponding to a value of $1.0 \times 10^5$ K/s. Several observations can be made from Fig. 14: (i) The $ftc$- and $ftc_2$-terms are mostly scattered up to $x \approx 7$ mm, corresponding to the approximate flame-base location as indicated by the temperature isolines. Downstream of this location, the two terms begin to exhibit more coherent structures, mostly congregated at the fuel-rich flame side. (ii) Both profiles appear well correlated with the flame-base and regions of high curvature. (iii) The $ftc$-term is larger than the $ftc_2$-term by as much as two orders of magnitude and is of comparable order to the heat-release rate. Figure 14 therefore provides a qualitative representation that demonstrates the presence of multidimensional effects, particularly that from the $ftc$-term, in this turbulent lifted hydrogen jet flame.

We will next examine the influence of multi-dimensional effects on the flamelets, starting with the regime diagram in Fig. 15. Note that (i) the $\kappa$-value in the regime diagrams is evaluated according to Eq. (58), but using the temperature $T$ instead of a reference species $Y_{i}$; and (ii) only points within the interval $0.1 < Z < 0.3$ are shown in the figure. This mixture fraction range corresponds to the region where the maximum temperature of the reacting flamelet population is found, thus serving as an appropriate measure for the reaction zone thickness on mixture-fraction space. In accordance with the regime diagram, we can discern that flamelets 3 and 4 cross over into the multi-dimensional flamelet regime III, while flamelets 1 and 2 occupy only the classical and curvature-affected flamelet regimes I and II. Therefore, it can be expected that only flamelets 3 and 4 will be affected by the $ftc_2$- and $ftc$-terms in their corresponding flamelet budgets, as illustrated in Fig. 16.

Figure 16 shows the $Y_{H_2O}, Y_{OH}, Y_{HO2}$, and $T$ budget analyses of the four flamelets that were evaluated in the regime diagram (cf. Fig. 15). Note that all budget plots have been normalized by their corresponding maximum absolute budget value, since we are concerned only by the relative orders of each contribution. In this regard, we also omitted negligible contributors such as $f_{\gamma}$ and $ft_{4}$ of the species and temperature in order to avoid data cluttering in the budget plots. Furthermore, terms such as $f_{4}, f_{c_2}$, and the $c_{y}$-component of $ft_{3}$ will not be relevant since the correction velocity [16] was not considered in the DNS; mass conservation was achieved by ensuring that $Y_{N_2} = 1 - \sum_{i=1}^{N_s} Y_{i}$, where $N_s$ is the number of species.

From Fig. 16, we can see that the budgets for $Y_{H_2O}, Y_{OH}$, and $T$ of flamelets 2, 3, and 4 have a multi-dimensional term $fc_2, ftc$ that is comparable to the classical flamelet terms. On the other hand, the tangential differential diffusion term $fc_2, ftc_2$ is generally of a smaller magnitude than the major flamelet terms. This trend is demonstrated also by the term’s physical interpretation in Fig. 14. The only exception is seen in the budgets for $Y_{H_2O}$ and...
For reference, the left-hand-side (LHS) of the flamelet Eqs. (56) and (57) is computed by adding all budget terms; this is represented by the black “Balance” line in the budget plots. In contrast to the laminar configuration, the balance of the four turbulent flamelets deviates significantly from zero, suggesting that the temporal term has a dominating influence on these flamelets. This can be attributed to the lifted flame configuration, since it is anticipated that the transient effects will reduce in magnitude for anchored flame configurations.

Combining the regime diagram (Fig. 15) and the budget analysis (Fig. 16), we find that the latter is generally consistent with the expectations based on the regime diagram, although there is admittedly a disagreement with regard to the importance of multi-dimensional effects along flamelet 2. However, we should recognize that the regime analysis is relevant only to the order-of-magnitude of the scaling parameters, $\varepsilon$ and $\nu$, and thus uncertainties within the same order are acceptable. Hence, it is remarkable that the budget plots and the regime diagram provide such a robust flame-regime characterization.

Considering the physical structures of the flamelets (cf. Fig. 13), it is possible to deduce a correlation between the relevance of the multi-dimensional term $f_{c1}$, $f_{tc}$ and the amount of corrugation and straining of the mixture fraction field. Specifically, we find that the probability of finding a flamelet in regime II and III is high in regions of high curvature (i.e. flamelets 2 and 3 based on concavity of the mixture fraction field) and/or low $\nu$ (i.e. flamelets 2 and 4 based on the spacing between mixture fraction isolines), and vice versa. To further support this observation, we construct the joint probability density function (PDF) of the two asymptotic scales, $\varepsilon$ and $\nu$, and scalar-dissipation rate $\chi$, as shown in Fig. 17. For this construction, all 2000 x 1600 points from the mid-plane were used. In Fig. 17, we present results for $P(\chi \leq 1, \varepsilon, \nu)$.
mixed according to the chemical explosive mode analysis by Lu et al. [22].

Sripakagorn et al. [22] demonstrate that the flamelet structure is retained; however, \( K_z \) requires consideration as an additional flamelet parameter. This regime is not present when considering equal diffusivities for mixture fraction, species and temperature.

In regime III, transport of species and temperature along mixture fraction isosurfaces becomes relevant. Due to the multi-dimensional character of this effect, the one-dimensionality of the flamelet structure becomes influential. In this regime a 1D flamelet structure is obtained; however, turbulent lifted hydrogen jet flame.

It is found that the relevance of tangential diffusion effects generally depends on the local curvature of the mixture fraction field, the scalar dissipation rate and the flame thickness \( \Delta Z_{fl} \).

By comparing relevant scales, a regime diagram is developed and three different flamelet regimes, that correspond to asymptotic distinguished limits, are identified. This diagram characterizes the relevance of the two competing mixing contributions due to curvature and scalar dissipation. In regime I, which is denoted as the classical flamelet regime, the flamelet solutions as derived by Peters [1] (Le = 1) and Pitsch and Peters [3] (Le \( \neq 1 \)) are valid. The regime II is called the curvature-affected flamelet regime, where differential diffusion of species and temperature along the flame-tangential direction becomes influential. In this regime a 1D flamelet structure is obtained; however, \( K_z \) requires consideration as an additional flamelet parameter. This regime is not present when considering equal diffusivities for mixture fraction, species and temperature.

In regime III, transport of species and temperature along mixture fraction isosurfaces becomes relevant. Due to the multi-dimensional character of this effect, the one-dimensionality of the flamelet structure becomes influential and regime III is denoted as multi-dimensional flamelet regime.

For verification of the asymptotic scaling three characteristic flamelets of the laminar co-flow flame were extracted. It is shown that the scalar fields of mixture fraction, species mass fraction and temperature exhibit the largest curvatures near the axis of the flame. Flamelet F1 is the flamelet along the jet axis and shows dominant multi-dimensional effects. This corresponds well to the characteristics of regime III. Flamelet F3 represents a flamelet solution at the flame base and exhibits dominant standard flamelet terms. This agrees well with regime I. Flamelet F2 represents a flamelet solution at the flame tip in a region of high curvature, showing a transition from regime I to III. The point of transition is found to be well predicted by the regime diagram obtained from the asymptotic scaling analysis.

The evaluation of the turbulent lifted hydrogen jet flame showed that the asymptotic scaling analysis is generally applicable to the turbulent flame regime. Using this diagnostic method, it was found that the probability of the multi-dimensional flamelet regime is as high as 21%, indicating that the multi-dimensional effect is a significant contribution in the flamelet-formulation. However, the budget analyses also clearly showed a strong presence of the temporal term, suggesting that the governing factor of the flamelet evolution may be unsteadiness, thereby complicating the problem with the necessity to consider transient effects on top of the multi-dimensional characteristic of the flamelets.

Nonetheless, the study carried out in this work shows that tangential diffusion effects are prevalent for certain laminar and turbulent flames. Neglecting tangential diffusion effects in these cases might introduce modeling errors with respect to flame structure and ignition characteristics in some regions of the flow. While certain model flames, e.g., laminar counterflow flames, can be modeled successfully using classical flamelet models, the additional terms accounting for tangential diffusion effects must be considered in the general case. The relevance of these effects can be assessed with the regime diagram derived in this work. Thus, the regime diagram can also be used as a diagnostic tool to evaluate the local model accuracy.

The application of flamelet models involves look-up of species and temperature data from a pre-tabulated chemistry solution. However, pre-processing the flamelet equations including tangential diffusion effects requires modeling since multi-dimensional terms are yet unenclosed. This closure remains subject to further research and is the criterion to successfully incorporate tangential diffusion effects into a multi-dimensional flamelet formulation.

5. Conclusions

In this work general flamelet equations including tangential diffusion effects are derived. The relevance of these terms is assessed by an asymptotic scaling and regime analysis and verified against budgets of the flamelet equations obtained from fully resolved CFD data of a laminar, non-premixed methane–air flame and a turbulent lifted hydrogen jet flame.

The most noticeable feature of Fig. 17 is the narrow band of high probability at around \( \varepsilon = 0.03 \). Even when its spreading is considered, the PDF always lies within 0.01 < \( \varepsilon < 0.06 \), indicating that \( 10^{-2} \) is an appropriate scale for \( \varepsilon \) when evaluated using Eq. (58).

In contrast, a shifting to larger \( \nu \) values can be seen as \( \chi \) increases, suggesting an inverse correlation between \( \varepsilon \) and \( v \). Recognizing that \( \chi = 2D_1\sqrt{\varepsilon^2} \) and \( v = \sqrt{\nu/\nu} \), we can infer that the inverse correlation also exists between the scalar-dissipation rate and the corrugation of the mixture-fraction field. Therefore, our previous statement with regards to the correlation between multi-dimensional effects and the level of curvature and strain of the mixture fraction field is hereby verified. It should be noted that this correlation is also supported by the results of the laminar case, which can be seen by comparing Figs. 10–12. To provide a quantitative measure of the relevance of multi-dimensional effects in the current turbulent flame configuration, we also evaluated the probability of regime III from the joint PDF to be approximately 21%, indicating that multi-dimensional effects are not negligible in the flamelet-formulation.

Indeed, the relevance of the \( f_{tc} \)-term should not be surprising. In a study on flame-reignition in a fully-developed homogeneous and isotropic turbulent field by Sripakagorn et al. [22], this term is responsible for interactions between extinguished and reacting flamelets. While flame quenching should be minimal in the current configuration due to the high co-flow temperature, the results of Sripakagorn et al. [22] demonstrate that the \( f_{tc} \)-term can provide a mechanism for flamelets to interact, thereby accounting for effects of flame-propagation. Such a capability will be applicable because the flamebase of the current configuration is largely premixed according to the chemical explosive mode analysis by Lu et al. [30].

The application of flamelet models involves look-up of species and temperature data from a pre-tabulated chemistry solution. However, pre-processing the flamelet equations including tangential diffusion effects requires modeling since multi-dimensional terms are yet unenclosed. This closure remains subject to further research and is the criterion to successfully incorporate tangential diffusion effects into a multi-dimensional flamelet formulation.

Fig. 17. Joint probability density function of \( \chi \) and \( \varepsilon \), constructed using points from the entire mid-plane at \( t = 1.3 \) ms, and cumulated within four \( \chi \) intervals, namely \( \chi < 1, 1 \leq \chi < 100, 100 \leq \chi < 1000, \) and \( \chi \geq 1000 \). For the probability-density-function construction, 50 bins have been used in each of the three coordinates.

\[
\mathcal{P}(1 \leq \chi \leq 100, \varepsilon, \nu), \mathcal{P}(100 \leq \chi \leq 1000, \varepsilon, \nu), \text{ and } \mathcal{P}(1000 \leq \chi, \varepsilon, \nu),
\]

where

\[
\mathcal{P}(\chi' \leq \chi \leq \chi', \varepsilon, \nu) = \int_{\chi'}^{\chi'} \mathcal{P}('\chi, \varepsilon, \nu) d\chi.
\]
Acknowledgments

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Appendix A. Lewis numbers when considering differential diffusion effects

The non-unity Lewis numbers have been chosen from a CFD calculation applying a full multi-component diffusion model [31] and the approximation [32]

\[ D_{jm} = \sum_{j=1}^{m} X_j D_{j} \]  

The Lewis numbers were averaged over a cross-section in the stoichiometric zone of the flame. These values were then used to carry out further CFD calculations, from which the numerical data for the budget analyses was obtained. For the case \( Le \neq 1 \) the species Lewis numbers are held constant as suggested by Pitsch and Peters [3] and are given in Table A.4. The Lewis number of the mixture fraction \( Le_z \) was set to unity.

Appendix B. Detailed derivation of the transformed species and temperature equations

In this section the detailed transformation of the non-dimensional Eqs. (21) and (22) into the \((\zeta, \eta, \tau, \xi_z)\)-system is given. Therefore, the transformation rules in Eqs. (37)–(39) and the scalings according to Eqs. (42), (45) and (47)–(51) are applied.

B.1. Transformation of the species Eq. (21)

Transforming the transient term yields

\[ \rho \frac{\partial Y_i}{\partial t} = \rho \frac{\partial Y_i}{\partial t} + \rho \frac{1}{v_i} \frac{\partial}{\partial \zeta} \frac{v_i}{\partial Y_i} \]  

(B.1)

The transformed convective terms read

\[ \rho v_i \frac{\partial Y_i}{\partial \eta} + \frac{3}{2} \rho \frac{\partial}{\partial \xi_z} Y_i \frac{\partial Y_i}{\partial \xi_z} = \frac{1}{v_i} \rho v_i \frac{\partial v_i}{\partial \eta} \frac{\partial Y_i}{\partial \xi_z} + \rho v_i \frac{\partial Y_i}{\partial \xi_z} \]  

(B.2)

\[ \text{Table A.4 Lewis numbers for all species.} \]

<table>
<thead>
<tr>
<th>Species</th>
<th>( Le )</th>
</tr>
</thead>
<tbody>
<tr>
<td>H</td>
<td>0.368</td>
</tr>
<tr>
<td>O</td>
<td>0.702</td>
</tr>
<tr>
<td>O2</td>
<td>0.839</td>
</tr>
<tr>
<td>OH</td>
<td>1.064</td>
</tr>
<tr>
<td>H2O</td>
<td>0.296</td>
</tr>
<tr>
<td>H2O2</td>
<td>0.177</td>
</tr>
<tr>
<td>H2</td>
<td>1.057</td>
</tr>
<tr>
<td>H2O</td>
<td>0.154</td>
</tr>
<tr>
<td>H2O2</td>
<td>1.405</td>
</tr>
<tr>
<td>CH4</td>
<td>1.267</td>
</tr>
<tr>
<td>C2H6</td>
<td>1.245</td>
</tr>
<tr>
<td>C3H8</td>
<td>1.278</td>
</tr>
<tr>
<td>C5H10</td>
<td>1.394</td>
</tr>
</tbody>
</table>

In the following the diffusive terms are transformed. The terms are numbered with respect to their appearance on the RHS of Eq. (21).

Diffusive term 1:

\[ \frac{\partial}{\partial \eta} \left( \rho D_i \frac{\partial Y_i}{\partial \eta} \right) = \frac{1}{\tau} \frac{\partial}{\partial \eta} \left( \rho D_i \frac{\partial Y_i}{\partial \tau} \right) + \frac{1}{\xi_z} \frac{\partial}{\partial \xi_z} \left( \rho D_i \frac{\partial Y_i}{\partial \xi_z} \right) \]

Diffusive term 2:

\[ \rho D_i \sum_{j=2}^{3} \frac{K_{j,2}}{\xi_z} \frac{\partial Y_j}{\partial \eta} = \rho D_i \sum_{j=2}^{3} \frac{1}{\tau} \frac{\partial}{\partial \eta} \left( \frac{K_{j,2}}{\xi_z} \frac{\partial Y_j}{\partial \tau} \right) + \rho D_i \sum_{j=2}^{3} \frac{1}{\xi_z} \frac{\partial}{\partial \xi_z} \left( \frac{K_{j,2}}{\xi_z} \frac{\partial Y_j}{\partial \xi_z} \right) \]

Diffusive term 3:

\[ \frac{\partial}{\partial \eta} \left( \rho D_i \frac{\partial Y_i}{\partial \eta} \right) = \frac{1}{\tau} \frac{\partial}{\partial \eta} \left( \rho D_i \frac{\partial Y_i}{\partial \tau} \right) + \frac{1}{\xi_z} \frac{\partial}{\partial \xi_z} \left( \rho D_i \frac{\partial Y_i}{\partial \xi_z} \right) \]

(B.3)

Diffusive term 2:

\[ \rho D_i \sum_{j=2}^{3} \frac{K_{j,2}}{v_i} \frac{\partial Y_j}{\partial \eta} = \rho D_i \sum_{j=2}^{3} \frac{1}{\tau} \frac{\partial}{\partial \eta} \left( \frac{K_{j,2}}{v_i} \frac{\partial Y_j}{\partial \tau} \right) + \rho D_i \sum_{j=2}^{3} \frac{1}{\xi_z} \frac{\partial}{\partial \xi_z} \left( \frac{K_{j,2}}{v_i} \frac{\partial Y_j}{\partial \xi_z} \right) \]

where Eqs. (9) and (48) were used. Diffusive term 3:

\[ \frac{3}{2} \frac{\partial}{\partial \eta} \left( \rho D_i \frac{\partial Y_i}{\partial \eta} \right) = \frac{1}{2} \frac{\partial}{\partial \eta} \left( \rho D_i \frac{\partial Y_i}{\partial \eta} \right) \]

(B.4)

Due to similarity the diffusive terms 4–6 are treated analogously to the diffusive terms 1–3.

Diffusive terms 7 and 8:

\[ \sum_{k=1}^{n} \frac{\partial}{\partial \eta} \left( \rho D_k \frac{\partial Y_k}{\partial \eta} \right) + \rho D_i \sum_{j=2}^{3} \frac{3}{2} \frac{\partial}{\partial \eta} \left( \rho D_j \frac{\partial Y_j}{\partial \eta} \right) \]

\[ = \sum_{k=1}^{n} \left[ \frac{1}{\tau} \frac{\partial D_k}{\partial \eta} \left( \frac{\partial Y_k}{\partial \tau} \right) + \frac{3}{\xi_z} \frac{\partial D_k}{\partial \xi_z} \left( \frac{\partial Y_k}{\partial \xi_z} \right) \right] \]

\[ + \rho D_i \sum_{j=2}^{3} \frac{3}{\tau} \frac{\partial}{\partial \eta} \left( \frac{K_{j,2}}{v_i} \frac{\partial Y_j}{\partial \tau} \right) + \rho D_i \sum_{j=2}^{3} \frac{1}{\xi_z} \frac{\partial}{\partial \xi_z} \left( \frac{K_{j,2}}{v_i} \frac{\partial Y_j}{\partial \xi_z} \right) \]

\[ + \rho D_i \sum_{j=2}^{3} \frac{3}{\tau} \frac{\partial}{\partial \eta} \left( \frac{K_{j,2}}{v_i} \frac{\partial Y_j}{\partial \tau} \right) + \rho D_i \sum_{j=2}^{3} \frac{1}{\xi_z} \frac{\partial}{\partial \xi_z} \left( \frac{K_{j,2}}{v_i} \frac{\partial Y_j}{\partial \xi_z} \right) \]

\[ + \rho D_i \sum_{j=2}^{3} \frac{3}{\xi_z} \frac{\partial D_j}{\partial \xi_z} \left( \frac{\partial Y_j}{\partial \xi_z} \right) \]

(B.5)

In Eq. (B.2) the first term on the RHS is replaced using the transport equation of the mixture fraction (Eq. (18), generalized for 3D case)

\[ \frac{1}{\tau} \frac{\partial}{\partial \eta} \left( \frac{\partial Y_i}{\partial \eta} \right) + \frac{1}{\xi_z} \frac{\partial}{\partial \xi_z} \left( \frac{\partial Y_i}{\partial \xi_z} \right) \]

(B.6)

Furthermore, \( \rho D_i \) is replaced by

\[ \rho D_i = \frac{Le_z}{Le} \rho D_z. \]

(B.7)

Using the definition of the scalar dissipation rate and (B.8) and collecting only leading order terms Eq. (B.3) is rewritten as
\[
\frac{\partial (\rho D_{1} \frac{\partial Y}{\partial r})}{\partial r} = \frac{1}{\varepsilon^2} \frac{\partial}{\partial r} \left( \frac{\rho Y}{\varepsilon^2} \right) \left( \frac{\partial Y}{\partial r} \right) + \frac{1}{\varepsilon} \frac{\partial}{\partial r} \left( \rho D_{1} \frac{\partial Y}{\partial r} \right) + \frac{1}{\varepsilon} \frac{\partial}{\partial r} \left( \frac{\rho Y}{\varepsilon} \frac{\partial Y}{\partial r} \right) + \frac{1}{\varepsilon} \frac{\partial}{\partial r} \left( \rho D_{1} \frac{\partial Y}{\partial r} \right) + \frac{1}{\varepsilon} \frac{\partial}{\partial r} \left( \frac{\rho Y}{\varepsilon} \frac{\partial Y}{\partial r} \right)
\]

(11)

Collecting the leading order terms of the transformed transient, convective and diffusive terms given above the transformed Eq. (21) reads

\[
\frac{\partial Y_i}{\partial r} + \frac{1}{\varepsilon} \frac{\partial}{\partial r} \left( \rho D_{1} \frac{\partial Y_i}{\partial r} \right) + \frac{1}{\varepsilon} \frac{\partial}{\partial r} \left( \rho Y_i \frac{\partial Y_i}{\partial r} \right) + \frac{1}{\varepsilon} \frac{\partial}{\partial r} \left( \rho D_{1} \frac{\partial Y_i}{\partial r} \right) + \frac{1}{\varepsilon} \frac{\partial}{\partial r} \left( \frac{\rho Y_i}{\varepsilon} \frac{\partial Y_i}{\partial r} \right)
\]

(12)

It can be shown that

\[
\frac{\partial}{\partial r} \left( \rho D_{1} \frac{\partial Y_i}{\partial r} \right) = \frac{1}{\varepsilon^2} \frac{\partial}{\partial r} \left( \rho Y_i \frac{\partial Y}{\partial r} \right) + \frac{1}{\varepsilon} \frac{\partial}{\partial r} \left( \rho D_{1} \frac{\partial Y}{\partial r} \right)
\]

(11)

where the definition of the scalar dissipation rate was used. Using the above relation in Eq. (10) yields the transformed Eq. (52).

B.2. Transformation of the temperature Eq. (22)

The transient and convective term of Eq. (22) are analogously transformed as the terms in Eqs. (1) and (2). In the following the transformation of the remaining terms of Eq. (22) is given. The terms are numbered according to their appearance on the RHS of the equation.

Term 1:

\[
\frac{1}{c_p} \frac{\partial}{\partial r} - \frac{1}{c_p} \frac{\partial}{\partial r} \left( \frac{\partial}{\partial r} \right)
\]

(12)

Term 2:

\[
\frac{\partial}{\partial r} \left( \frac{\partial}{\partial r} \right) = \frac{1}{c_p} \frac{\partial}{\partial r} \left( \frac{\partial}{\partial r} \right) + \frac{1}{c_p} \frac{\partial}{\partial r} \left( \frac{\partial}{\partial r} \right) + \frac{1}{c_p} \frac{\partial}{\partial r} \left( \frac{\partial}{\partial r} \right) + \frac{1}{c_p} \frac{\partial}{\partial r} \left( \frac{\partial}{\partial r} \right) + \frac{1}{c_p} \frac{\partial}{\partial r} \left( \frac{\partial}{\partial r} \right)
\]

(13)

where the definition of the scalar dissipation rate was used and the thermal diffusivity \( \lambda/c_p \) was replaced by \( \lambda/c_p = \rho D_T \).

(14)

These two relations are further used in the transformation of the remaining terms. The transformation of term 3 reads:

\[
\frac{\partial}{\partial r} \left( \rho D_{1} \frac{\partial T}{\partial r} \right) = \frac{1}{\varepsilon^2} \frac{\partial}{\partial r} \left( \rho Y_i \frac{\partial Y}{\partial r} \right) + \frac{1}{\varepsilon} \frac{\partial}{\partial r} \left( \rho D_{1} \frac{\partial Y}{\partial r} \right) + \frac{1}{\varepsilon} \frac{\partial}{\partial r} \left( \frac{\rho Y_i}{\varepsilon} \frac{\partial Y}{\partial r} \right)
\]

(11)

Collecting all leading order terms and using the Eqs. (18) (generalized for 3D case) and (11) yields

\[
\frac{\lambda}{c_p} \sum_{\beta=2}^{3} \frac{\partial T}{\partial r} = d_{e} \frac{\partial T}{\partial r} + \frac{1}{c_p} \frac{\partial}{\partial r} \left( \frac{\partial}{\partial r} \right)
\]

(13)
\[
\rho \frac{\partial T}{\partial t} + \sum_{\beta=2}^{3} \rho u_{\beta} \frac{\partial T}{\partial x_{\beta}} = \frac{1}{2 \epsilon} \left( \frac{\rho T}{\bar{\epsilon}^2} \right) \frac{\partial \bar{\epsilon}}{\partial x_{\beta}} + \frac{1}{2 \epsilon} \left( \frac{\rho_2 T}{\epsilon} \right) \frac{1}{c_p} \frac{\partial T}{\partial c_p} \frac{\partial T}{\partial c_p}
\]

\[
- \frac{1}{2} \frac{\rho T}{\bar{\epsilon}^2} \frac{1}{c_p} \frac{\partial T}{\partial c_p} \sum_{i=1}^{n} (c_{p_{\beta}} - c_{p_{\beta}}) \frac{\partial T}{\partial x_{\beta}}
\]

\[
+ \frac{1}{4} \frac{1}{\epsilon^2} \left[ 2 \rho T \frac{\partial T}{\partial c_p} + (Le_2 - 1) \frac{1}{\epsilon^2} \frac{\partial T}{\partial c_p} \frac{\partial T}{\partial c_p} \right] + \frac{1}{\epsilon^2} \frac{1}{2 \epsilon} \left( \frac{\rho T}{\epsilon} \right) \frac{1}{c_p} \frac{\partial T}{\partial c_p} \sum_{i=1}^{n} h_{\beta} m_{\beta}.
\]

(B.20)

A backward transformation analogously to the species equation in Section 3.3 yields the flamelet equation for the temperature, Eq. (57).

### Appendix C. Definition of second derivatives with respect to \(s_p\)

The flamelet equations contain second derivatives with respect to \(s_p\). When projecting the \(Z\) isosurface into the \(x_1-x_2\) plane it appears as a \(X\) isoline and \(s_p\) is defined as the coordinate measured along the \(Z\) isoline. For notational convenience \(x_1\) and \(x_2\) are denoted as \(x \) and \(s\) for the following two-dimensional considerations.

The \(Z\) isoline is defined as

\[ dZ = 0 = \frac{\partial Z}{\partial x} dx + \frac{\partial Z}{\partial y} dy. \]

Rewriting the above expression the differential equation of the \(Z\) isoline is obtained

\[ dx = \frac{\partial Z}{\partial y} dy. \]

From Eq. (C.1) the coordinate \(s\) is defined as

\[ ds = \left( \frac{dx}{dy} \right) - ds = |ds| = \sqrt{dx^2 + dy^2}. \]

First derivatives of an arbitrary scalar quantity \(\phi\) can be expressed as

\[ \frac{\partial \phi}{\partial s} = \frac{ds}{|ds|} \cdot \nabla \phi = \frac{1}{\sqrt{dx^2 + dy^2}} \left( \frac{dx}{dy} \right) \cdot \left( \frac{\partial \phi}{\partial x} \frac{\partial \phi}{\partial y} \right) = \frac{\partial \phi}{\partial x} \frac{\partial x}{\partial y} + \frac{\partial \phi}{\partial y} \frac{\partial y}{\partial x}. \]

(C.4)

where Eq. (C.2) was used. Consequently, second derivatives are obtained from

\[ \frac{\partial^2 \phi}{\partial s^2} = \frac{\partial}{\partial s} \left( \frac{\partial \phi}{\partial s} \right) = \frac{\frac{\partial^2 \phi}{\partial x^2}}{\sqrt{\left( \frac{\partial x}{\partial y} \right)^2 + \left( \frac{\partial y}{\partial x} \right)^2}}. \]

(C.5)

For the axial flamelet as investigated by Xu et al. [12] \(x\) is the coordinate measured along the axis and first order derivatives with respect to \(y\) are zero due to axial symmetry. It follows that \(\partial \phi/\partial x = 0\) and it can be shown from Eq. (C.5) that

\[ \frac{\partial^2 \phi}{\partial s^2} = \frac{\frac{\partial^2 \phi}{\partial x^2}}{\left( \frac{\partial x}{\partial y} \right)^2} \left( \frac{\partial x}{\partial y} \right) \left( \frac{\partial y}{\partial x} \right). \]

(C.6)

It has to be noted that first order derivatives with respect to \(y\) may not be dropped before differentiation because second derivatives with respect to \(y\) are non-zero. Applying the transformation as reported by Xu et al. [12] \(\partial \phi/\partial \epsilon\) can be transformed

\[ \frac{\partial \phi}{\partial \epsilon} = \frac{\partial \phi}{\partial \epsilon} \frac{\partial \phi}{\partial \epsilon}. \]

(C.7)

Thus, using Eq. (C.7) and the definition of the curvature analogously to Eq. (10) the LHS of Eq. (C.6) can be reformulated as

\[ \frac{\partial^2 \phi}{\partial s^2} = \sqrt{\frac{\nabla Z}{2D}} \frac{\kappa\phi}{\nabla Z} \]

where \(\kappa\phi\) is defined for a 2D case as [12]

\[ \kappa\phi = -\frac{\phi_\phi}{|\nabla\phi|^3}. \]

When replacing \(\phi\) by the temperature or the species mass fraction of a product species, \(\phi\) will exhibit a maximum within the reaction zone and the value of \(\partial \phi/\partial \epsilon\) turns to zero. Nevertheless, the quantity \(\partial^2 \phi/\partial s^2\) shows finite values at this point as \(\kappa\phi\) tends to infinity which can be readily seen from Eq. (C.9).

### C.1. Extension to three-dimensional space

Since the turbulent lifted hydrogen jet flame is simulated in the three-dimensional Cartesian space, a volumetric equivalent of Eq. (C.5) is needed to evaluate the second derivative with respect to \(s_p\). Such extension is made possible by the following decomposition of the Eulerian Laplacian of a generic scalar \(\phi\):

\[ \nabla \phi = \nabla_2 \phi + \nabla_1 \phi, \]

(C.10)

where the \(\nabla_2\) and \(\nabla_1\) operators can be interpreted as the diffusion of \(\phi\) in directions normal and tangential to the mixture-fraction isosurface. The definitions of the two operators are given as follows:

\[ \nabla_2 \phi = \nabla Z \cdot \nabla \phi, \]

(C.11a)

\[ \nabla_1 \phi = \frac{1}{\epsilon} \nabla_2 \phi - \kappa_\phi \nabla Z \cdot \nabla \frac{\partial \phi}{\partial \epsilon}, \]

(C.11b)

where \(\nabla Z = \nabla Z/|\nabla Z|\) is the normalized gradient of mixture-fraction.

Another decomposition of the Eulerian Laplacian can be made by retaining the outer divergerence operator and splitting the inner scalar gradient according to the flamelet transformation \((\nabla Z) = \nabla Z (\nabla Z) / \partial \epsilon + \nabla Z (\nabla Z) / \partial Z\):

\[ \nabla_1 \phi = \nabla \cdot (\nabla_1 \phi) = \nabla \cdot \left( \nabla Z \frac{\partial \phi}{\partial \epsilon} + \nabla_2 \phi \right), \]

(C.12)

where the first term on the RHS can be further described, using Eq. (C.11a), as:

\[ \nabla \cdot (\nabla Z \frac{\partial \phi}{\partial \epsilon}) = \nabla \cdot (\nabla Z) \frac{\partial \phi}{\partial \epsilon} + \nabla \cdot \nabla Z \cdot \frac{\partial (\nabla \phi)}{\partial \epsilon}, \]

\[ = \nabla \cdot (\nabla Z) \cdot \nabla_1 \phi + \nabla \cdot (\nabla Z) \cdot \nabla_2 \phi, \]

\[ + \nabla \cdot (\nabla Z) \cdot \nabla_1 \phi + \nabla \cdot (\nabla Z) \cdot \nabla_2 \phi, \]

\[ = -\kappa_\phi \nabla Z \cdot \nabla_2 \phi + \nabla \cdot (\nabla Z) \cdot \nabla_2 \phi, \]

(C.13)
One caveat to Eq. (C.13) is its implicit assumption of an orthonormal coordinate system such that the derivative with respect to Z (i.e. ∂/∂Z) is equal to ⟨∇Z⟩−1⟨n2 · ∇⟩. Essentially, this assumption asserts that the covariant vector for the local curvilinear transformation is ⟨∇Z⟩−1n2.

As the current evaluation does indeed enforce an orthonormal coordinate system, the Eqs. (C.10), (C.11b), and (C.13) can be combined to give:

\[ \sum_{\beta=2}^{3} \frac{\partial^3 \phi}{\partial \beta^2} = \nabla \cdot (\nabla_{\beta} \phi). \]  

(C.14)

This final form is simple, yet insightful because it provides a physical interpretation to the second derivative with respect to s2 as the divergence of the tangential components of the scalar gradient. Furthermore, Eq. (C.14) shows that the second derivative with respect to s2 has been rightfully categorized as a tangential diffusion effect. It should be noted that this form of the tangential diffusion term is general to different quantities since no assumption of the scalar has been made.

Interestingly, Eq. (C.14) can be shown to be consistent with the two-dimensional Eq. (C.5). In order to prove this point, the final form (C.14) has to be expanded in terms of the x- and y-coordinates:

\[ \frac{\partial^2 \phi}{\partial x^2} = \nabla \cdot (\nabla_{\phi} \phi) = \left( \frac{\partial \phi}{\partial x} \right)^2 + \left( \frac{\partial \phi}{\partial y} \right)^2 - \frac{\partial^2 \phi}{\partial x \partial y} - \frac{\partial^2 \phi}{\partial y \partial x} + \frac{\partial^2 \phi}{\partial x \partial y} + \frac{\partial^2 \phi}{\partial y \partial x} - \frac{\partial^2 \phi}{\partial x \partial y} - \frac{\partial^2 \phi}{\partial y \partial x}. \]  

(C.15)

For the axial flamelet as investigated by Xu et al. [12], where x is aligned with the mixture-fraction gradient (i.e. ⟨∇Z⟩ = 0) and ∂/∂y = 0 due to axial symmetry, Eq. (C.15) can be reduced to:

\[ \frac{\partial^2 \phi}{\partial x^2} = \frac{(\partial \phi)^2}{\partial x^2} - \frac{(\partial \phi)^2}{\partial y^2} - \frac{2 \partial \phi}{\partial y} \frac{\partial^2 \phi}{\partial x \partial y} - \frac{2 \partial \phi}{\partial x} \frac{\partial^2 \phi}{\partial y \partial x} + \frac{\partial^2 \phi}{\partial x \partial y} + \frac{\partial^2 \phi}{\partial y \partial x}, \]

which is precisely Eq. (C.6).

### Appendix D. Consistencies with previous works

To date, several studies that investigated the effects of tangential diffusion can be found [4,12,13,22]. Although these studies are referring to the same subject of interest, the sets of notation differ widely. In this section it is the intention to show consistencies of previous contributions in comparison to the findings of this work.

#### D.1. Consistency with Williams [21]

The first evaluation refers to a generic flamelet equation given by Williams [21] obtained for Le = 1. Species and temperature equations are transformed into an orthogonal coordinate system, where Z is one of the coordinates and the other two are distances along surfaces of constant Z, which precisely correspond to the two coordinates s2 and s3 introduced in Section 3. The species equation reads

\[ \rho \frac{\partial Y_i}{\partial t} + \rho \mathbf{v} \cdot \nabla Y_i = m_i + \rho D |\nabla Z|^2 \nabla^2 Y_i + \nabla \cdot (\rho D \nabla Y_i) - \rho D \nabla (\ln |\nabla Z|) \cdot \nabla Y_i, \]  

(D.1)

where \( \mathbf{v} \) denotes components in the iso-mixture fraction plane, namely

\[ \mathbf{v}_s = \mathbf{v} - \mathbf{a} \frac{\nabla Z}{|\nabla Z|} = \left( \begin{array}{c} v_x \\ v_y \\ v_z \end{array} \right), \]

(D.2)

and \( \nabla \) is the gradient operator in s2 and s3. The identities of the gradient and the divergence operator \( \nabla \) can also be defined by [13]

\[ \nabla \phi = \phi - \nabla Z \frac{\partial \phi}{\partial Z}, \]

(D.3a)

\[ \nabla \cdot \mathbf{a} = \nabla \cdot \mathbf{a} - \nabla Z \frac{\partial \mathbf{a}}{\partial Z}. \]  

(D.3b)

where \( \phi \) denotes a scalar quantity and \( \mathbf{a} \) is a vector. The transformed gradient in the \( (Z, s_2, s_3) \) coordinate system reads

\[ \nabla \phi = \left( \begin{array}{c} \frac{\partial \phi}{\partial Z} \\ \frac{\partial \phi}{\partial s_2} \\ \frac{\partial \phi}{\partial s_3} \end{array} \right). \]

(D.4)

where components with respect to \( \xi_1 - \xi_3 \) where neglected, since these terms cancel to leading order in the vicinity of the point of reference \( \text{ref} \) (see Appendix B). With this, it can be shown that the gradient \( \nabla_{\beta} \phi \) equals to

\[ \nabla_{\beta} \phi = \left( \begin{array}{c} \frac{\partial \phi}{\partial x} \\ \frac{\partial \phi}{\partial y} \\ \frac{\partial \phi}{\partial z} \end{array} \right). \]

(D.5)

Further, the transformed Laplacian equals to

\[ \nabla^2 \phi = \frac{\partial^2 \phi}{\partial x^2} + \frac{\partial^2 \phi}{\partial y^2} + \frac{\partial^2 \phi}{\partial z^2} - \frac{\partial^2 \phi}{\partial x \partial y} - \frac{\partial^2 \phi}{\partial y \partial x} + \frac{\partial^2 \phi}{\partial x \partial z} + \frac{\partial^2 \phi}{\partial y \partial z} + \frac{\partial^2 \phi}{\partial z \partial x} - \frac{\partial^2 \phi}{\partial x \partial z} + \frac{\partial^2 \phi}{\partial z \partial y} - \frac{\partial^2 \phi}{\partial y \partial z}. \]

(D.6)

With the previous relations, it is shown that Eq. (D.1) is fully consistent with the species equation derived in this work. Simplifying Eq. (56) for Le = 1 yields

\[ \rho \frac{\partial Y_i}{\partial t} + \sum_{\beta=2}^{3} \rho \mathbf{v}_s \cdot \frac{\partial Y_i}{\partial s_\beta} = m_i + \frac{\rho}{2} \frac{\partial^2 Y_i}{\partial Z^2} + \rho \sum_{\beta=2}^{3} \frac{\partial^2 Y_i}{\partial s_\beta^2}. \]  

(D.7)
It is readily shown that the terms on the left hand side and the first two terms on the right hand side in Eq. (D.1) are consistent with the corresponding terms in Eq. (D.7). Expanding the third term on the right hand side of Eq. (D.1) yields

\[
\nabla \cdot (\rho D \nabla Y_i) = \nabla \cdot (\rho D \nabla Y_i) - \nabla Z \cdot \frac{\partial}{\partial Z} (\rho D \nabla Y_i) \\
= \nabla \rho D \cdot \nabla Y_i + \rho D \nabla Z \cdot \frac{\partial}{\partial Z} Y_i - \rho D \nabla Z \cdot \frac{\partial}{\partial Z} (\rho D \nabla Y_i) \\
= \sum_{p=2}^{3} \frac{\partial \rho D}{\partial x_p} \frac{\partial Y_i}{\partial x_p} + \rho D \nabla Z \cdot \frac{\partial}{\partial Z} (\rho D \nabla Y_i),
\]

(D.8)

where Eqs. (19) and (4.4)-(6.6) were used. The first term on the right hand side is a higher order term (see Appendix B) and the second term is equal to the multi-dimensional term in Eq. (D.7). It was shown by Xuan et al. [13, see Appendix B] that the third term on the right hand side exactly balances the fourth term in Eq. (D.1). From the term-by-term comparison it is concluded that Eq. (D.7) is fully consistent with Eq. (D.1). Thus, the flamelet model proposed in this work is equivalent for \( \text{Le} = 1 \) to the formulation suggested by Williams [21].

D.2. Consistency with Xu et al. [12]

Xu et al. [12] investigated the axisymmetric flamelet of a reactive coal particle in a convectiv flow. Since a similar notation is chosen, species and temperature flamelet equations of their work are readily related to the Eqs. (56) and (57). Due to symmetry they could introduce simplifications that do not hold when considering the general case. In Appendix C it is shown that the multi-dimensional terms derived in this work can be simplified to the corresponding term of Xu et al. Furthermore, additional terms in Eqs. (56) and (57) originate from variable Lewis numbers that were held constant by Xu et al. Hence, the Eqs. (56) and (57) can be considered the general form of the flamelet equations proposed by Xu et al. [12].

D.3. Consistency with Xuan et al. [13]

In a very recent work Xuan et al. [13] investigated and modeled curvature effects in the laminar flamelet formulation. Their species flamelet equation is given as:

\[
\rho \frac{\partial Y_s}{\partial t} = \left( \frac{\rho Y_s}{\text{Le}} \right) \frac{\partial^2 Y_s}{\partial Z^2} + \frac{\partial^2 Y_s}{\partial Z^2} + \bar{m}_s \\
+ \sum_{p=2}^{3} \left( \frac{\rho Y_s}{\text{Le}_s} \frac{\partial^2 Y_s}{\partial x_p^2} + \nabla \cdot (\rho D_s \nabla Y_s) \right) \\
+ \frac{\rho D_s}{\text{Le}_s} (\nabla Z \cdot \nabla Y_s) \frac{\partial^2 Y_s}{\partial Z^2} + \frac{1}{\text{Le}_s} \sum_{p=2}^{3} \nabla \cdot (\rho D_s \nabla Y_s) \frac{\partial^2 Y_s}{\partial Z^2}.
\]

(D.9)

where non-unity, but constant Lewis number and constant molecular weight \( W \) have been assumed. For simplicity, the correction velocity term is neglected in this comparison. On the other hand, the species flamelet equation that is employed in the current work can be written, once again omitting the correction velocity term, as:

\[
\rho \frac{\partial Y_s}{\partial t} = \left( \frac{\rho Y_s}{\text{Le}} \right) \frac{\partial^2 Y_s}{\partial Z^2} + \bar{m}_s \\
+ \sum_{p=2}^{3} \left( \frac{\rho Y_s}{\text{Le}_s} \frac{\partial^2 Y_s}{\partial x_p^2} + \nabla \cdot (\rho D_s \nabla Y_s) \right) \\
+ \frac{\rho D_s}{\text{Le}_s} (\nabla Z \cdot \nabla Y_s) \frac{\partial^2 Y_s}{\partial Z^2} + \frac{1}{\text{Le}_s} \sum_{p=2}^{3} \nabla \cdot (\rho D_s \nabla Y_s) \frac{\partial^2 Y_s}{\partial Z^2}.
\]

(D.10)

Due to the attempt to maintain the simpler Eulerian form in Eq. (D.9), the equivalence between the two equations is not apparent by merely stating them. In order to draw the connection, expansions of certain terms are necessary and presented in the following (using the notation according to Eqs. (C.10), (C.11), and (C.14)):

\[
\nabla \cdot (\rho D_s \nabla Y_s) = \frac{1}{4} \left( \frac{\partial^2 Y_s}{\partial Z^2} + \rho D_s \frac{\partial \log \rho D_s}{\partial Z} \right) - \rho D_s \frac{\partial Y_s}{\partial Z}.
\]

(D.11a)

\[
\sum_{p=2}^{3} \frac{\partial^2 Y_s}{\partial x_p^2} = \nabla \cdot (\nabla Y_s) = \left( \nabla \cdot \frac{\partial Y_s}{\partial Z} \right) \frac{\partial Z}{\partial Z} + \left( \nabla Z \cdot \frac{\partial Y_s}{\partial Z} \right) + \nabla Z \cdot \frac{\partial^2 Y_s}{\partial Z^2} \\
+ \frac{\partial \partial Z}{\partial Z} \frac{\partial Y_s}{\partial Z} + \nabla Z \cdot \frac{\partial^2 Y_s}{\partial Z^2} + \frac{\partial \partial Z}{\partial Z} \frac{\partial Y_s}{\partial Z}.
\]

(D.11b)

Substituting Eq. (D.11) into Eq. (D.9), the species equation by Xuan et al. [13] can then be rewritten as:
\[
\frac{\partial Y_x}{\partial t} + \rho \sum_{j=2}^{3} \left( \frac{\partial Y_x}{\partial Z_2} \left( \frac{\partial Z_2}{\partial t} + \mathbf{u} \cdot \nabla Z_2 \right) \right) = \frac{1}{\rho} \frac{\partial}{\partial Z_2} \left( \frac{\partial Y_x}{\partial Z_2} \right) \frac{\partial Z_2}{\partial t} + m_x.
\]

which can be viewed as the counterpart of Eq. (D.10) in scenarios where the terms in Eq. (D.15) significantly dominate over the other budget terms such as the multi-dimensional terms. Under such cases, it is reasonable to further approximate the coefficient of the curvature based terms as one, since it typically has a unity order-of-magnitude (i.e., \(1/(Le - 1) \approx 0(1))\). In other words, one can regard the formulation of Kortschik et al. [4] as the leading-order approximation of the more general Eq. (D.10), which is still remarkable, given that it was the first attempt to capture tangential effects on a flamelet.

D.6. Summary

To summarize, the Eqs. (56) and (57) are demonstrated to be general and consistent with previous investigations. For their comprehensiveness, these flamelet formulations can be regarded as a standard equation set to guide modeling efforts. Future work to extend the flamelet model should gear towards accounting for every budget term in the equations. In this way, the capabilities of the flamelet concept can then be fully exploited, possibly allowing for the accurate representation of: (i) geometrical effects such as curvature of the mixture-fraction field as studied separately by Kortschik et al. [4] and Xuan et al. [13] and (ii) flamelet interaction through tangential diffusion as analyzed by Sripakagorn et al. [22].

Appendix E. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.combustflame.2014.11.016.

References


